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(54) Title: METHOD OF MAKING WASHABLE, DRYABLE ELASTIC ARTICLES (57) Abstract <p>The present invention relates to a method of making improved polyolefinic elastic articles from cured, irradiated or cross-linked amorphous ethylene interpolymers. In particular, the invention relates to a method of making a shaped article (for example, film or fiber) characterized by improved elevated temperature elasticity as well as washability and dryability. The inventive elastic article comprises a substantially cured, irradiated, cross-linked (or curable, irradiated or cross-linkable) homogeneously branched ethylene interpolymer characterized as having a density less than 0.90 g/cm³ and containing at least one nitrogen-containing stabilizer. The improved elastic article of the present invention is particularly suitable for use in applications where good elasticity must be maintained at elevated temperatures and after laundering such as, for example, elastic waist bands of undergarments and other clothing.</p>		

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METHOD OF MAKING WASHABLE, DRYABLE ELASTIC ARTICLES

The present invention relates to a method of making
5 improved polyolefinic elastic articles from cured, irradiated
or crosslinked amorphous ethylene interpolymers. In
particular, the invention relates to a method of making a
shaped article (for example, film or fiber) characterized by
improved elevated temperature elasticity as well as
10 washability and dryability. The inventive elastic article
comprises a substantially cured, irradiated, or crosslinked
(or curable, irradiated or crosslinkable) homogeneously
branched ethylene interpolymer characterized as having a
density less than 0.90 g/cm^3 and containing at least one
15 nitrogen-containing stabilizer. The improved elastic article
of the present invention is particularly suitable for use in
applications where good elasticity must be maintained at
elevated temperatures and after laundering such as, for
example, elastic waist bands of undergarments and other
20 clothing.

Materials with excellent stretchability and elasticity
are needed to manufacture a variety of disposal and durable
articles such as, for example, incontinence pads, disposable
diapers, training pants, clothing, undergarments, sports
25 apparel, automotive trim, weather-stripping, gaskets, and
furniture upholstery. Stretchability and elasticity are
performance attributes which can, for example, function to
effectuate a closely conforming fit to the body of the wearer
or to the frame of the item. While numerous materials are
30 known to exhibit excellent stress-strain properties and
elasticity at room temperatures, it is often desirable for
elastic materials to provide a conforming or secure fit

during repeated use, extensions and retractions at elevated temperatures such as at body temperatures or in automobile interiors during summer months. Maintaining tight tolerances throughout temperature cycles are also instances where elevated temperature elasticity is important. Further, where an elastics material is employed in clothing or garment items, the material must maintain its integrity and elastic performance after laundering.

Disposable elastic articles are typically elastic composite materials prepared from a combination of polymer film, fibers, sheets and/or absorbent materials as well as a combination of fabrication technologies. Whereas elastic fibers can be prepared by well known processes such as spun bonding, melt blowing, melt spinning and continuous filament wounding techniques, the film and sheet forming processes typically involve known extrusion and coextrusion techniques, for example, blown film, cast film, profile extrusion, injection molding, extrusion coating, and extrusion sheeting.

Conversely, durable elastic articles are often molded or profile items such as, for example, automotive door and window trim, clothing waist band threads or strips, and building weather-stripping. Such durable articles can be made by well known molding, thermoforming and profile technologies.

A material is typically characterized as elastic when it is characterized as having a high percent elastic recovery (that is, a low percent permanent set) after application of a biasing force. Ideally, elastic materials are characterized by a combination of three, temperature independent properties, that is, a low percent permanent set, a low stress or load at strain, and a low percent stress or load relaxation. That is, there should be at low to elevated

service temperatures (1) a low stress or load requirement to stretch the material, (2) no or low relaxing of the stress or unloading once the material is stretched, and (3) complete or high recovery to original dimensions after the stretching, 5 biasing or straining is discontinued.

Lycra® is the trademark of Dupont Fibers for its elastic spandex fibers. The U.S. International Trade Commission defines spandex fiber as a manufactured fiber in which the fiber-forming substance is a long-chain synthetic polymer 10 comprised of at least 85 percent segmented polyurethane. Lycra is known to exhibit nearly ideal, temperature independent elastic properties rendering it very suitable for use in garments, sports apparel and swimsuits. However, one significant shortcoming of Lycra is it typically exhibits 15 fair to poor elevated temperature serviceability and washability. Similar to ordinary uncrosslinked polyolefin-based elastic materials, Lycra articles tend to lose their integrity and shape and elastic properties when subjected to elevated service temperatures such as during laundering and 20 drying. Another major shortcoming of Lycra is its cost. That is, Lycra tends to be extremely cost prohibitive for a many of applications.

Elastic materials such as films, strips, coating, ribbons and sheet comprising at least one substantially 25 linear ethylene polymer are disclosed in U.S. Patent No. 5,472,775 to Obijeski et al. However, U.S. Patent No. 5,472,775 does not disclose the performance of these materials at elevated temperatures (that is, at temperatures above room temperature), nor their performance after 30 laundering.

WO 94/25647 discloses elastic fibers and fabrics made from homogeneously branched substantially linear ethylene

polymers. The fibers are said to possess at least 50 percent recovery (that is, less than or equal 50 percent permanent set) at 100 percent strain. However, there is no disclosure in WO 94/25647 regarding the elasticity of these fibers at
5 elevated temperatures or the effects of laundering on these fibers.

WO 95/29197 discloses curable, silane-grafted substantially ethylene polymers which are useful in wire and cable coatings, weather-stripping, and fibers. In the
10 Examples, inventive samples include fibers comprising silane-grafted substantially ethylene polymers having densities of 0.868 g/cm³ and 0.870 g/cm³. The inventive examples are shown to exhibit improved elastic recovery at elevated
15 temperatures. However, there is no disclosure in WO 95/29197 regarding the percent stress or load relaxation performance at elevated temperatures for these silane-crosslinked fibers, nor is there any disclosure as to washability.

U.S. Patent No. 5,324,576 discloses an elastic nonwoven web of microfibers of radiation crosslinked ethylene/alpha
20 olefin copolymers, preferably having a density less than 0.9 g/cm³. In the examples set forth in U.S. Patent No. 5,324,576, ethylene polymers having polymer densities greater than or equal to 0.871 g/cm³ are subjected to electron beam radiation. However, there is no disclosure regarding the
25 elastic performance of these radiated polymers at elevated temperatures, nor is there any disclosure regarding their resistance to washing and drying.

U.S. Patent No. 5,525,257 to Kurtz et al. discloses that low levels of irradiation of less than 2 megarads of Ziegler
30 catalyzed linear low density ethylene polymer results in improved stretchability and bubble stability without measurable gelation. However, '257 provides no disclosure

respecting the elasticity and/or washability at elevated temperatures.

U.S. Patent No. 4,957,790 to Warren discloses the use of pro-rad compounds and irradiation to prepare heat-shrinkable linear low density polyethylene films having an increased orientation rate during fabrication. In the examples provided therein, Warren employs Ziegler catalyzed ethylene polymers having densities greater than or equal to 0.905 g/cm³.

Various compounds are disclosed in the art and/or sold commercially as high temperature stabilizers and antioxidants. However, the criteria employed to distinguish these compounds as stabilizers and antioxidants typically relates to their ability to resistance yellowing, crosslinking and/or the ill-effects of irradiation (for example, gamma irradiation for purposes of sterilization).

In other instances, different types of stabilizers are equated to one another or are said to perform comparably. For example, it is known that hindered phenolic stabilizers (for example, Irganox[®] 1010 supplied by Ciba-Geigy) can be as effective as hindered amine stabilizers (for example, Chimassorb[®] 944 supplied by Ciba-Geigy), and vice versa. In a product brochure entitled, "Chimassorb 944FL: Hindered Amine Light Stabilizer Use and Handling", printed 1996, Ciba-Geigy states Chimassorb 9944 "gives long-term heat stability to polyolefins by a radical trapping mechanism similar to that of hindered phenols."

Further, there is some belief that there is no universally effective stabilizer for polymers as the definition for stability inevitably varies with each application. In particular, there is no effective stabilizer

for washable, high temperature serviceable polyolefinic elastic materials.

In general, stabilizers are known to inhibit crosslinking. In regard to crosslinking generally, there are several disclosures relating to radiation resistant (for example, gamma and electron beam) polymer compositions comprising amine stabilizers. Such disclosures typically teach relatively high levels of amine stabilizer (for example, greater than or equal to 0.34 weight percent) are required where inhibition of crosslinking, discoloration and other undesirable irradiation effects is desired. Another examples include stabilized disposal nonwoven fabrics (see, for example, U.S. Patent No. 5,200,443) and stabilized molding materials (for example, syringes). Gamma sterilization resistant fibers, including amine coatings and the use of hybrid phenolic/amine stabilizers are also known. See, for example, U.S. Patent No. 5,122,593 to Jennings et al.

Stabilized polyethylene compositions with improved resistance to oxidation and improved radiation efficiency are also known. M. Iring et al. in "The Effect of the Processing Steps on the Oxidative Stability of Polyethylene Tubing Crosslinked by Irradiation", Die Angew. Makromol. Chemie, Vol. 247, pp. 225-238 (1997) teach that amine stabilizers are more effective towards inhibiting electron-beam irradiation effects (that is, provide better resistance against oxidation) than hindered phenols.

WO 92/19993 and U.S. Patent No. 5,283,101 discloses launderable retroreflective appliqués comprised of a multicomponent binder composition consisting of an electron-beam curable elastomer, crosslinker(s), and coupling agent(s) and optional colorants, stabilizers, flame retardants and

flow modifiers. The allegedly inventive appliquéés are said to be capable of withstanding ordinary household washing conditions as well as more stringent industrial washings without loss of retroreflectiveness. Illustrative examples of electron-beam curable elastomers of the binder are said to be "chlorosulfonated polyethylenes, ethylene copolymers comprising at least about 70 weight percent of polyethylene such as ethylene/vinyl acetate, ethylene/acrylate, and ethylene/acrylic acid, and poly(ethylene-co-propylene-co-diene) ("EPDM") polymers." Optional stabilizers are described to be "thermal stabilizers and antioxidants such as hindered phenols and light stabilizers such as hindered amines or ultraviolet stabilizers". Although there is an equating of the suitability or effectiveness of hindered phenols to hindered amines in the descriptions of WO 92/19993 and U.S. Patent No. 5,283,101, no stabilizer of any kind is exemplified in the provided examples. Further, although the appliqué can employ polymers that are described as "highly flexible" before and after electron-beam curing, neither the selected polymers nor the appliqué itself are described as "elastic". That is, a material can be highly flexible yet nonelastic as the terms "nonelastic" and "elastic" are defined herein below. However, the reverse is not true; elastic materials are characterized as having a high degree of flexibility (that is, Young's Modulus of less than 10,000 psi (68.9 Mpa) where lower modulus means more flexibility).

Although there is an abundance of art related to elastic ethylene polymer articles, including articles comprising curable, radiated and/or crosslinked ethylene polymers, and an abundance of art related to stabilized compositions and articles, there is no known disclosure of a polyolefinic elastic material with effective additive stabilization

wherein the stabilization does not inhibit the desirable effects of irradiation and/or crosslinking (designed to impart elevated temperature elasticity and an increased melting point) and yet does inhibit the loss of elastic integrity (that is, scission) when the material is subjected to a detergent washing and drying at elevated temperatures. Further, in another product brochure entitled, "Stabilization of Adhesives and Their Components", pp. 8-9 (1994), Ciba-Geigy, a premier stabilizer supplier, states that scission occurring in elastomeric materials (for example, styrene-isoprene-styrene block copolymers) at elevated temperatures above 70°C is not readily controlled by the use of antioxidants.

As such, there is a present need for cost-effective, stable elastic articles having good elasticity at elevated temperatures as well as good washability and dryability. That is, there is a need for elastic articles which retain their shapes under strain at elevated temperature (for example, greater than or equal to 125°C). There is also a need for a method of making elastic articles having good elasticity at elevated temperatures and good wash/dry stability. We have discovered that these and other objects can be completely met by the invention herein described.

Summary of the Invention

We have discovered that elastic articles comprising curable, irradiated and/or crosslinkable ethylene interpolymers characterized by a polymer density of less than 0.90 g/cm³ at 23°C and at least one nitrogen-containing stabilizer exhibit excellent elasticity at room temperature and at elevated temperatures as well as excellent wash and dry stability. According to the broad aspect of the

invention, there is provided a method of making a shaped curable, irradiated or crosslinkable article comprising at least one homogeneously branched ethylene interpolymer, which comprises ethylene interpolymerized with at least one other
5 monomer and characterized as having (before being shaped, grafted, cured, irradiated, or crosslinked) a polymer density of less than 0.90 g/cm³ at 23°C, and at least one nitrogen-containing stabilizer.

Another aspect of the invention is a method of making a
10 shaped and cured, irradiated or crosslinked article comprising at least one homogeneously branched ethylene interpolymer, which comprises ethylene interpolymerized with at least one other monomer and characterized as having (before being shaped, grafted, cured, irradiated, or
15 crosslinked) a polymer density of less than 0.90 g/cm³ at 23°C, and at least one nitrogen-containing stabilizer.

A third aspect of the invention is a method of making an elastic article comprising the steps of:

(a) providing at least one homogeneously branched
20 ethylene interpolymer having a density of less than 0.90 g/cm³ at 23°C having at least 0.1 weight percent of at least one nitrogen-containing stabilizer therein,

(b) fabricating or shaping the article from the interpolymer, and

25 (c) after the fabrication or shaping, subjecting the article to heat and/or ionizing radiation, wherein the article is characterized as having:

(i) a percent permanent set of less than or equal 25 at
23°C and 200 percent strain when measured at a 2 mil
30 (102 mm) thickness using an Instron tensiometer after being shaped and cured, irradiated or crosslinked,

(ii) a percent stress relaxation of less than or equal 25 at 23°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using a Instron tensiometer after being shaped and cured, irradiated or crosslinked, and

(iii) a percent stress relaxation of less than or equal 55 at 38°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using an Instron tensiometer after.

A fourth aspect of the invention is a method of making an elastic article wherein the steps further comprises incorporating a pro-rad crosslink additive into the interpolymer.

A fifth aspect of the invention is a method of making a curable elastic article comprising the steps of:

(a) providing at least one homogeneously branched ethylene interpolymer characterized as having a density at 23°C less than 0.90 g/cm³ and comprising at least 0.1 weight percent of at least one nitrogen-containing stabilizer incorporated therein,

(b) preparing a melt of the stabilized interpolymer of (a);

(c) mixing into the melt of (b) from 0.5 to 5 phr of a silane crosslinker (parts of silane crosslinker per hundred parts interpolymer) while the crosslinker is at an ambient temperature between 0 and 30°C; and

(d) subjecting the melt mixture of (c) to ionizing energy or contacting the melt mixture of (c) with at least one free radical initiator to graft at least 50 weight percent, based on the total weight of the crosslinker and the

interpolymer, of the silane crosslinker to the stabilized interpolymer.

Preferably, the article is fabricated or shaped using an extrusion technique (that is, the method consists of melting the interpolymer) such as, for example, a fiber melt spinning, fiber melt blowing, film blowing, cast film, injection molding, or rotomolding technique, and is permitted to cool or is quenched to ambient temperature (that is, permitted to substantially solidify) before the application or exposures to (additional) heat, ionizing radiation and/or moisture.

In a preferred embodiment of the invention, the at least one homogeneously branched ethylene interpolymer is a substantially linear ethylene interpolymer. In another preferred embodiment, the ionizing radiation is provided by electron beam irradiation. In a third preferred embodiment, the at least one nitrogen-containing stabilizer is a hydroquinoline, diphenylamine or substituted piperidine.

We discovered that there is a subset of ethylene polymers which provide completely unexpected elastic performance results when cured, radiated and/or crosslinked. In particular, we found for a broad range of polymer densities, curing, radiation and/or crosslinking can dramatically decrease percent permanent set performance (that is, improve elasticity or elastic recovery) and have no substantial effects on ambient percent stress or load relaxation performance. However, while tending to adversely affect (that is, increase) or have no affect on percent stress or load relaxation at elevated temperatures for polymer having densities equal to or greater than 0.865 g/cm^3 , surprisingly curing, radiation and crosslinking decreases (that is, improves) the elevated temperature percent stress

or load relaxation performance of ethylene interpolymer having a polymer density less than 0.865 g/cm³ or a DSC crystallinity at 23°C less than 8.5 weight percent. That is, curing, radiating and/or crosslinking is an effective means for providing elastic materials and articles characterized as having excellent elevated temperature stress relaxation characteristics.

Not only is the dramatically different response to irradiation or crosslinking surprisingly in itself, these results are surprising for another reasons as well. For example, these results are surprisingly and unexpected because at a density less than 0.90 g/cm³, ethylene interpolymers are already substantially amorphous. That is, a cross-over or transition in elastic performance attributable to curing, radiation and/or crosslinking would ordinarily be expected to relate to the amorphosity of the polymer; however, according to hexane extraction data at 50°C, determined according to the Food and Drug Administration (FDA) test method set forth under 21 37 C.F.R. § § 177.1520 (d) (3) (ii), ethylene polymers are substantially amorphous at a density of 0.89 g/cm³ and below. Given such small differences in amorphosity or crystallinity, dramatic elasticity differences in response to irradiation or crosslinking simply would not ordinarily be expected.

As another surprise, we discovered that the incorporation of at least one nitrogen-containing stabilizer imparts excellent laundering characteristics to the elastic article. This discovery is surprising and unexpected because the stabilizer does not inhibit or interfere with effective curing, radiation effects, crosslinking or crosslinking effects (and as such permits substantial melting point increases, that is, from less than 75°C to greater than

125°C), yet inhibits melting and flowing (that is, scission) from occurring at substantially elevated temperatures (for example, 133°C) in a wash/extend dry testing.

The washing and drying performance results of the inventive article are also surprising for at least one other reason. That is, the effectiveness of the at least one nitrogen-containing stabilizer is unexpected because in ordinary stabilization tests (for example, inhibition of yellowing) nitrogen-containing stabilizers perform comparable to phenolic stabilizers, yet phenolic stabilizers do not inhibit melting and flowing in wash/dry testing.

FIG. 1 is a plot of percent stress relaxation at 23°C versus megarads of electron beam radiation for Inventive Examples 1 and 2 and comparative examples 3, 4 and 5.

FIG. 2 is a plot of percent permanent set at 23°C versus megarads of electron beam radiation for Inventive Examples 1 and 2 and comparative examples 3, 4 and 5.

FIG. 3 is a plot of percent stress relaxation at 38°C versus megarads of electron beam radiation for Inventive Example 1 and comparative example 4.

The term "elastic" as used herein refers to a material having a permanent set of less than 60 percent, especially less than or equal to 25 percent (that is, especially greater than or equal to 87.5 percent recovery) at 200 percent strain (wherein 200 percent strain is, for example, stretching a 1 inch (2.5 cm) article to a final dimension of 3 inches). Elastic materials are more than simply highly flexible as in addition to having a Young's Modulus of less than 10,000 psi (68.9 Mpa), they are defined a low percent permanent set at 200 percent strain. Elastic materials are also referred to in the art as "elastomers" and "elastomeric".

The term "nonelastic" as used herein means the material

or article is not elastic as defined herein (that is, the martial or article has a percent permanent set greater than 25).

Elastic materials and articles include, the cured,
5 radiated and/or crosslinked ethylene interpolymer itself as well as, but are not limited, a fiber, film, strip, tape, ribbon, sheet, coating, and molding comprised of the cured, radiated and/or crosslinked ethylene interpolymer. Preferred elastic articles are fiber and film.

10 The term "radiated" or "irradiated" as used herein means the ethylene polymer, the shaped ethylene interpolymer or the article comprised of the ethylene polymer was subjected to at least 3 megarads (or the equivalent thereof) of ionizing energy whether or not there was a measurable decrease in
15 percent xylene extractables (that is, increase in insoluble gel). That is, substantial crosslinking may not result from the irradiation.

The terms "crosslinked" and "substantially crosslinked" as used herein mean the ethylene polymer, the shaped ethylene
20 interpolymer or the article comprised of the ethylene polymer is characterized as having xylene extractables of less than 85 weight percent, preferably less than or equal to 75 weight percent, more preferably less than or equal to 70 weight percent, where xylene extractables are determined in
25 accordance with ASTM D-2765.

The terms "cured" and "substantially cured" as used herein means the ethylene interpolymer, the shaped ethylene interpolymer or the article comprised of the ethylene interpolymer was subjected or exposed to a treatment which
30 induced crosslinking. As used herein, the terms relate to ethylene interpolymers comprising a grafted silane.

The terms "curable" and "crosslinkable" as used herein

mean the ethylene interpolymer, the shaped ethylene interpolymer or the article comprised of the ethylene interpolymer is not crosslinked and has not been subjected or exposed to treatment which induces crosslinking although the
5 ethylene interpolymer, the shaped ethylene interpolymer or the article comprised of the ethylene interpolymer comprises additive(s) or functionality which will effectuate crosslinking upon subjection or exposure to such treatment.

The term "pro-rad additive" as used herein means a
10 compound which is not activated during normal fabrication or processing of the homogeneously branched ethylene interpolymer, however can be activated by the application of temperatures (heat) substantial above normal fabrication or processing temperatures and/or by ionizing energy to
15 effectuate some measurable gelation or preferably, substantial crosslinking.

The term "homofil" as used herein refers to fiber which has a single polymer region or domain and does not have any other distinct polymer regions (as do bicomponent fibers).

20 The term "meltblown" is used herein in the conventional sense to refer to fibers formed by extruding a molten thermoplastic polymer composition through a plurality of fine, usually circular, die capillaries as molten threads or filaments into converging high velocity gas streams (for
25 example, air) which function to attenuate the threads or filaments to reduced diameters. Thereafter, the filaments or threads are carried by the high velocity gas streams and deposited on a collecting surface to form a web of randomly dispersed meltblown fibers with average diameters generally
30 smaller than 10 microns.

The term "spunbond" is used herein in the conventional sense to refer to fibers formed by extruding a molten

thermoplastic polymer composition as filaments through a plurality of fine, usually circular, die capillaries of a spinneret with the diameter of the extruded filaments then being rapidly reduced and thereafter depositing the filaments
5 onto a collecting surface to form a web of randomly dispersed spunbond fibers with average diameters generally between 7 and 30 microns.

The term "nonwoven" as used herein and in the conventional sense means a web or fabric having a structure
10 of individual fibers or threads which are randomly interlaid, but not in an identifiable manner as is the case for a knitted fabric. The elastic fiber of the present invention can be employed to prepare nonwoven fabrics as well as composition structures comprises elastic nonwoven fabric in
15 combination with nonelastic materials.

The term "conjugated" refers to fibers which have been formed from at least two polymers extruded from separate extruders but meltblown or spun together to form one fiber. Conjugated fibers are sometimes referred to in the art as
20 multicomponent or bicomponent fibers. The polymers are usually different from each other although conjugated fibers may be monocomponent fibers. The polymers are arranged in substantially constantly positioned distinct zones across the cross-section of the conjugated fibers and extend
25 continuously along the length of the conjugated fibers. The configuration of conjugated fibers can be, for example, a sheath/core arrangement (wherein one polymer is surrounded by another), a side by side arrangement, a pie arrangement or an "islands-in-the sea" arrangement. Conjugated fibers are
30 described in U.S. Patent No. 5,108,820 to Kaneko et al.; U.S. Patent No. 5,336,552 to Strack et al.; and U.S. Patent No. 5,382,400 to Pike et al. The elastic fiber of the present

invention can be in a conjugated configuration, for example, as a core or sheath, or both.

The at least one homogeneously branched ethylene interpolymer to be irradiated, cured and/or crosslinked has a
5 density at 23°C less than 0.90 g/cm³, preferably less than or equal to 0.865 g/cm³, more preferably in the range of 0.865 g/cm³ to 0.855 g/cm³, as measured in accordance with ASTM D792. At densities higher than 0.90 g/cm³, the interpolymer is not substantially amorphous or elastic, even at room
10 temperature. Further, while at densities of equal to or less than 0.87 g/cm³, the benefits of effective stabilization can be realized, densities of equal to or less than 0.865 g/cm³ are preferred because the desired improvement in high temperature elastic performance (especially, a low percent
15 stress or load relaxation) is obtained.

Preferably, the homogeneously branched ethylene interpolymer is characterized as having a DSC crystallinity of less than or equal to 8.3 weight percent, more preferably less than or equal to 8 weight percent and most preferably
20 less than or equal to 6 percent.

Preferably, the homogeneously branched ethylene interpolymer is characterized as having a melt index less than 10 g/10 minutes, as determined in accordance with ASTM D-1238, Condition 190°C/2.16 kilogram (kg).

25 The irradiated, cured and/or crosslinked article of the present invention is characterized as having a percent permanent set of less than 60 at 23°C, preferably less than or equal to 25 at 23°C, more preferably less than or equal to 20 and most preferably less than or equal to 15 at 23°C and 38°C
30 and 200 percent strain when measured at a 2 mil (102 mm) thickness using an Instron tensiometer.

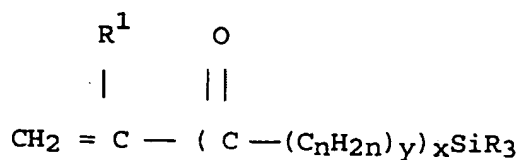
The irradiated, cured and/or crosslinked article of the present invention (and is characterized as having a percent stress relaxation of less than or equal to 25 at 23°C and 200 percent strain and less than or equal to 55, preferably less than or equal to 50, more preferably less than or equal to 30, most preferably less than or equal to 20 at 38°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using a Instron tensiometer.

Irradiation may be accomplished by the use of high energy, ionizing electrons, ultra violet rays, X-rays, gamma rays, alpha particles, protons, and beta particles and combination thereof. However, electron-beam irradiation is preferred. The irradiation is preferably carried out at a dosage up to 70 megarads, more preferably between 3 megarads to 35 megarads, most preferably between 4 to 30 megarads. Further, the irradiation can be carried out conveniently at room temperature, although higher and lower temperatures, for example 0°C to 60°C, may also be employed. Preferably, the irradiation is carried out after shaping or fabrication of the article. Also, in a preferred embodiment, the homogeneously branched ethylene interpolpolymer with a pro-rad additive incorporated therein is irradiated with electron beam radiation at 8 to 20 megarads.

The electron-beam irradiation source can be any suitable electron-beam generator. For example, suitable electron-beam irradiation equipment is available from Energy Services, Inc. Wilmington, Mass. with capabilities of at least 100KeV and at least 5 Kw. The voltage can be adjusted to appropriate levels such as, for example, 100,000, 300,000, 1,000,000 or 2,000,000 or 3,000,000 or 6,000,000 or higher or lower. Many other apparati for irradiating polymeric materials are also known in the art..

The ethylene homogeneously branched interpolymer may be crosslinked or cured by first grafting a silane onto its polymer backbone and thereafter subjecting or exposing the silane grafted ethylene interpolymer to water or atmospheric moisture. Preferably, the silane grafted ethylene polymer is subjected or exposed to water or atmospheric moisture after a shaping or fabrication operation.

Suitable silanes for silane crosslinking of the ethylene interpolymer include those of the general formula



in which R' is a hydrogen atom or methyl group; x and y are 0 or 1 with the proviso that when x is 1, y is 1; n is an integer from 1 to 12 inclusive, preferably 1 to 4, and each R independently is a hydrolyzable organic group such as an alkoxy group having from 1 to 12 carbon atoms (for example, methoxy, ethoxy, butoxy), aryloxy group (for example, phenoxy), araloxy group (for example, benzyloxy), aliphatic acyloxy group having from 1 to 12 carbon atoms (for example, formyloxy, acetyloxy, propanoyloxy), amino or substituted amino groups (alkylamino, arylamino), or a lower alkyl group having 1 to 6 carbon atoms inclusive, with the proviso that not more than one of the three R groups is an alkyl.

Suitable silanes may be grafted to a suitable ethylene polymer by the use of a suitable quantity of organic peroxide, either before or during a shaping or fabrication operation. However, preferably, the silane is grafted onto the ethylene interpolymer before shaping or fabrication operations. In any case, the curing or crosslinking reaction

takes place following the shaping or fabrication operation by reaction between the grafted silane groups and water. The water permeating into the bulk polymer from the atmosphere or from a water bath or "sauna". The phase of the process
5 during which the crosslinks are created is commonly referred to as the "cure phase" and the process itself is commonly referred to as "curing".

Any silane that will effectively graft to and crosslink the ethylene interpolymer can be used in the present
10 invention. Suitable silanes include unsaturated silanes that comprise an ethylenically unsaturated hydrocarbyl group, such as a vinyl, allyl, isopropenyl, butenyl, cyclohexenyl or γ -(meth)acryloxy allyl group, and a hydrolyzable group, such as, for example, a hydrocarbyloxy, hydrocarbonyloxy, or
15 hydrocarbylamino group. Examples of hydrolyzable groups include methoxy, ethoxy, formyloxy, acetoxy, propionyloxy, and alkyl or arylamino groups. Preferred silanes are the unsaturated alkoxy silanes which can be grafted onto the polymer. These silanes and their method of preparation are
20 more fully described in U.S. Patent No. 5,266,627 to Meverden, et al. Vinyl trimethoxy silane, vinyl triethoxy silane, γ -(meth)acryloxy propyl trimethoxy silane and mixtures of these silanes are the preferred silane crosslinkers for use in this invention. If a filler (for example, calcium
25 carbonate, talc, mica, silica (for example, SiO_2 , clay, and aluminum trihydrate,) is present, then preferably the crosslinker includes vinyl triethoxy silane.

The amount of silane crosslinker used in the present invention can vary widely depending several factors such as
30 the silane itself, processing conditions, grafting efficiency, organic peroxide selection, the ultimate application, and similar factors. However, typically at

least 0.5, preferably at least 0.7, parts per hundred resin (phr) is used. Considerations of convenience and economy are usually the two principal limitations on the maximum amount of silane crosslinker used, and typically the maximum amount of silane crosslinker does not exceed 5, preferably it does not exceed 2, phr. As used in parts per hundred resin or phr, "resin" means the ethylene interpolymer.

The silane crosslinker is grafted to the ethylene interpolymer by any conventional method, typically in the presence of a free radical initiator for example, peroxides and azo compounds, or by ionizing radiation, etc. A suitable grafting method is disclosed in WO 95/29197.

However, for efficient silane grafting, organic initiators are preferred, such as any one of the peroxide initiators, for example, dicumyl peroxide, di-tert-butyl peroxide, t-butyl perbenzoate, benzoyl peroxide, cumene hydroperoxide, t-butyl peroctoate, methyl ethyl ketone peroxide, 2,5-dimethyl-2,5-di(t-butyl peroxy)hexane, lauryl peroxide, and tert-butyl peracetate. A suitable azo compound is azobisisobutyl nitrite. The amount of initiator can vary, but it is typically present in an amount of at least 0.04, preferably at least 0.06, phr. Typically, the initiator does not exceed 0.15, preferably it does not exceed 0.10, phr. The ratio of silane crosslinker to initiator also can vary widely, but the typical crosslinker:initiator ratio is between 10:1 to 30:1, preferably between 18:1 and 24:1.

While any conventional method can be used to graft the silane crosslinker to the substantially linear ethylene polymer, one preferred method is blending the two with the initiator in the first stage of a reactor extruder, such as a Buss kneader. The grafting conditions can vary, but the melt temperatures are typically between 160 and 260 C, preferably

between 190 and 230 C, depending upon the residence time and the half life of the initiator.

Cure is can be promoted with a crosslinking catalyst, and any catalyst that will provide this function can be used. Suitable catalysts generally include organic bases, carboxylic acids, and organometallic compounds and combinations thereof including organic titanates and complexes or carboxylates of lead, cobalt, iron, nickel, zinc and tin. Representative catalyst include, for example, but is not limited to, dibutyltindilaurate, dioctyltinmaleate, dibutyltindiacetate, dibutyltindiocetoate, stannous acetate, stannous octoate, lead naphthenate, zinc caprylate, and cobalt naphthenate. Tin carboxylate, especially dibutyltindilaurate and dioctyltinmaleate, are particularly effective for this invention. The catalyst (or mixture of catalysts) is present in a catalytic amount, typically between 0.015 and 0.035 phr.

Representative pro-rad additives include, but are not limited to, azo compounds, organic peroxides and polyfunctional vinyl or allyl compounds such as, for example, triallyl cyanurate, triallyl isocyanurate, pentaerthritol tetramethacrylate, glutaraldehyde, ethylene glycol dimethacrylate, diallyl maleate, dipropargyl maleate, dipropargyl monoallyl cyanurate, dicumyl peroxide, di-tert-butyl peroxide, t-butyl perbenzoate, benzoyl peroxide, cumene hydroperoxide, t-butyl peroctoate, methyl ethyl ketone peroxide, 2,5-dimethyl-2,5-di(t-butyl peroxy)hexane, lauryl peroxide, tert-butyl peracetate, azobisisobutyl nitrite and combination thereof. Preferred pro-rad additives for use in the present invention are compounds which have polyfunctional (that is, at least two) moieties such as C=C, C=N or C=O.

At least one pro-rad additive can be introduced to the homogeneously branched ethylene interpolpolymer by any method known in the art. However, preferably the pro-rad additive(s) is introduced via a masterbatch concentrate
5 comprising the same or different base resin as the ethylene interpolpolymer. Preferably, the pro-rad additive concentration for the masterbatch is relatively high for example, 25 weight percent (based on the total weight of the concentrate).

The at least one pro-rad additive is introduced to the
10 homogeneously branched ethylene polymer in any effective amount. Preferably, the at least one pro-rad additive introduction amount is from 0.001 to 5 weight percent, more preferably from 0.005 to 2.5 weight percent and most preferably from 0.015 to 1 weight percent (based on the total
15 weight of the ethylene interpolpolymer).

The term "polymer", as used herein, refers to a polymeric compound prepared by polymerizing monomers, whether of the same or a different type. As used herein, generic term "polymer" embraces the terms "homopolymer," "copolymer,"
20 "terpolymer" as well as "interpolymer."

The term "interpolymer", as used herein refers to polymers prepared by the polymerization of at least two different types of monomers. As used herein the generic term "interpolymer" includes the term "copolymers" (which is
25 usually employed to refer to polymers prepared from two different monomers) as well as the term "terpolymers" (which is usually employed to refer to polymers prepared from three different types of monomers).

The term "homogeneously branched ethylene polymer" is
30 used herein in the conventional sense to refer to an ethylene interpolpolymer in which the comonomer is randomly distributed within a given polymer molecule and wherein substantially all

of the polymer molecules have the same ethylene to comonomer molar ratio. The term refers to an ethylene interpolymer that are manufactured using so-called homogeneous catalyst systems known in the art such Ziegler vanadium, hafnium and zirconium catalyst systems and metallocene catalyst systems (for example, a constrained geometry catalyst systems) as described by Elston in U.S. Patent No. 3,645,992; Stevens et al. in U.S. Patent No. 5,064,802 and EP 0 416 815 A2; Canich in U.S. Patent No. 5,026,798 and U.S. Patent No. 5,055,438; Parikh et al. in WO 93/13143; and Kolthammer et al. in WO 94/17112.

Homogeneously branched ethylene polymers for use in the present invention can be also described as having less than 15 weight percent, preferably less 10 weight percent, more preferably less than 5 and most preferably zero (0) weight percent of the polymer with a degree of short chain branching less than or equal to 10 methyls/1000 carbons. That is, the polymer contains no measurable high density polymer fraction (for example, there is no fraction having a density of equal to or greater than 0.94 g/cm³), as determined, for example, using a temperature rising elution fractionation (TREF) technique and infrared or ¹³C nuclear magnetic resonance (NMR) analysis.

Preferably, the homogeneously branched ethylene polymer is characterized as having a narrow, essentially single melting TREF profile/curve and essentially lacking a measurable high density polymer portion, as determined using a temperature rising elution fractionation technique (abbreviated herein as "TREF").

The composition distribution of an ethylene interpolymer can be readily determined from TREF as described, for example, by Wild et al., Journal of Polymer Science, Poly.

Phys. Ed., Vol. 20, p. 441 (1982), or in U.S. Patent No. 4,798,081; U.S. Patent No. 5,008,204; or by L. D. Cady, "The Role of Comonomer Type and Distribution in LLDPE Product Performance," SPE Regional Technical Conference, Quaker Square Hilton, Akron, Ohio, October 1-2, pp. 107-119 (1985).

The composition (monomer) distribution of the interpolymer can also be determined using ^{13}C NMR analysis in accordance with techniques described in U.S. Patent No. 5,292,845; U.S. Patent No. 4,798,081; U.S. Patent No. 5,089,321 and by J. C. Randall, Rev. Macromol. Chem. Phys., C29, pp. 201-317.

In analytical temperature rising elution fractionation analysis (as described in U.S. Patent No. 4,798,081 and abbreviated herein as "ATREF"), the film or composition to be analyzed is dissolved in a suitable hot solvent (for example, trichlorobenzene) and allowed to crystallize in a column containing an inert support (stainless steel shot) by slowly reducing the temperature. The column is equipped with both a refractive index detector and a differential viscometer (DV) detector. An ATREF-DV chromatogram curve is then generated by eluting the crystallized polymer sample from the column by slowly increasing the temperature of the eluting solvent (trichlorobenzene). The ATREF curve is also frequently called the short chain branching distribution (SCBD) or composition distribution (CD) curve, since it indicates how evenly the comonomer (for example, octene) is distributed throughout the sample in that as elution temperature decreases, comonomer content increases. The refractive index detector provides the short chain distribution information and the differential viscometer detector provides an estimate of the viscosity average molecular weight. The composition distribution and other compositional information can also be

determined using crystallization analysis fractionation such as the CRYSTAF fractional analysis package available commercially from PolymerChar, Valencia, Spain.

Preferred homogeneously branched ethylene polymers (such as, but not limited to, substantially linear ethylene polymers) have a single melting peak between -30 and 150°C, as determined using differential scanning calorimetry (DSC), as opposed to traditional Ziegler polymerized heterogeneously branched ethylene polymers (for example, LLDPE and ULDPE or VLDPE) which have two or more melting points.

The single melting peak is determined using a differential scanning calorimeter standardized with indium and deionized water. The method involves 5-7 mg sample sizes, a "first heat" to 180°C which is held for 4 minutes, a cool down at 10°C/min. to -30°C which is held for 3 minutes, and heat up at 10°C/min. to 150°C to provide a "second heat" heat flow vs. temperature curve from which the melt peak(s) is taken. Total heat of fusion of the polymer is calculated from the area under the curve.

The homogeneously branched ethylene polymers for use in the invention can be either a substantially linear ethylene polymer or a homogeneously branched linear ethylene polymer.

The term "linear" as used herein means that the ethylene polymer does not have long chain branching. That is, the polymer chains comprising the bulk linear ethylene polymer have an absence of long chain branching, as in the case of traditional linear low density polyethylene polymers or linear high density polyethylene polymers made using Ziegler polymerization processes (for example, U.S. Patent No. 4,076,698 (Anderson et al.)), sometimes called heterogeneous polymers. The term "linear" does not refer to bulk high pressure branched polyethylene, ethylene/vinyl acetate

copolymers, or ethylene/vinyl alcohol copolymers which are known to those skilled in the art to have numerous long chain branches.

The term "homogeneously branched linear ethylene
5 polymer" refers to polymers having a narrow short chain branching distribution and an absence of long chain branching. Such "linear" uniformly branched or homogeneous polymers include those made as described in U.S. Patent No. 3,645,992 (Elston) and those made using so-called single site
10 catalysts in a batch reactor having relatively high ethylene concentrations (as described in U.S. Patent No. 5,026,798 (Canich) or in U.S. Patent No. 5,055,438 (Canich)) or those made using constrained geometry catalysts in a batch reactor also having relatively high olefin concentrations (as
15 described in U.S. Patent No. 5,064,802 (Stevens et al.) or in European Patent No. 0 416 815 A2 (Stevens et al.)).

Typically, homogeneously branched linear ethylene polymers are ethylene/ α -olefin interpolymers, wherein the α -olefin is at least one C_3 - C_{20} α -olefin (for example,
20 propylene, 1-butene, 1-pentene, 4-methyl-1-pentene, 1-hexene, 1-heptene, and 1-octene) and preferably the at least one C_3 - C_{20} α -olefin is 1-butene, 1-hexene, 1-heptene or 1-octene. Most preferably, the ethylene/ α -olefin interpolymers is a copolymer of ethylene and a C_3 - C_{20} α -olefin, and especially an
25 ethylene/ C_4 - C_8 α -olefin copolymer such as an ethylene/1-octene copolymer, ethylene/1-butene copolymer, ethylene/1-pentene copolymer or ethylene/1-hexene copolymer.

Suitable homogeneously branched linear ethylene polymers for use in the invention are sold under the designation of
30 TAFMER by Mitsui Chemical Corporation and under the designations of EXACT and EXCEED resins by Exxon Chemical Company.

The term "substantially linear ethylene polymer" as used herein means that the bulk ethylene polymer is substituted, on average, with 0.01 long chain branches/1000 total carbons to 3 long chain branches/1000 total carbons (wherein "total carbons" includes both backbone and branch carbons). Preferred polymers are substituted with 0.01 long chain branches/1000 total carbons to 1 long chain branches/1000 total carbons, more preferably from 0.05 long chain branches/1000 total carbons to 1 long chain branched/1000 total carbons, and especially from 0.3 long chain branches/1000 total carbons to 1 long chain branches/1000 total carbons.

As used herein, the term "backbone" refers to a discrete molecule, and the term "polymer" or "bulk polymer" refers, in the conventional sense, to the polymer as formed in a reactor. For the polymer to be a "substantially linear ethylene polymer", the polymer must have at least enough molecules with long chain branching such that the average long chain branching in the bulk polymer is at least an average of from 0.01/1000 total carbons to 3 long chain branches/1000 total carbons.

The term "bulk polymer" as used herein means the polymer which results from the polymerization process as a mixture of polymer molecules and, for substantially linear ethylene polymers, includes molecules having an absence of long chain branching as well as molecules having long chain branching. Thus a "bulk polymer" includes all molecules formed during polymerization. It is understood that, for the substantially linear polymers, not all molecules have long chain branching, but a sufficient amount do such that the average long chain branching content of the bulk polymer positively affects the melt rheology (that is, the shear viscosity and melt fracture

properties) as described herein below and elsewhere in the literature.

Long chain branching (LCB) is defined herein as a chain length of at least one (1) carbon less than the number of
5 carbons in the comonomer, whereas short chain branching (SCB) is defined herein as a chain length of the same number of carbons in the residue of the comonomer after it is incorporated into the polymer molecule backbone. For example, a substantially linear ethylene/1-octene polymer has
10 backbones with long chain branches of at least seven (7) carbons in length, but it also has short chain branches of only six (6) carbons in length.

Long chain branching can be distinguished from short chain branching by using ^{13}C nuclear magnetic resonance (NMR)
15 spectroscopy and to a limited extent, for example, for ethylene homopolymers, it can be quantified using the method of Randall, (Rev. Macromol.Chem. Phys., C29 (2&3), p. 285-297). However as a practical matter, current ^{13}C nuclear magnetic resonance spectroscopy cannot determine the length
20 of a long chain branch in excess of about six (6) carbon atoms and as such, this analytical technique cannot distinguish between a seven (7) carbon branch and a seventy (70) carbon branch. The long chain branch can be as long as about the same length as the length of the polymer backbone.

25 Although conventional ^{13}C nuclear magnetic resonance spectroscopy cannot determine the length of a long chain branch in excess of six carbon atoms, there are other known techniques useful for quantifying or determining the presence of long chain branches in ethylene polymers, including
30 ethylene/1-octene interpolymers. For example, U.S. Patent No. 4,500,648 teaches that long chain branching frequency (LCB) can be represented by the equation $\text{LCB} = b/M_w$ wherein b is the

weight average number of long chain branches per molecule and M_w is the weight average molecular weight. The molecular weight averages and the long chain branching characteristics are determined by gel permeation chromatography and intrinsic
5 viscosity methods, respectively.

Two other useful methods for quantifying or determining the presence of long chain branches in ethylene polymers, including ethylene/1-octene interpolymers are gel permeation chromatography coupled with a low angle laser light
10 scattering detector (GPC-LALLS) and gel permeation chromatography coupled with a differential viscometer detector (GPC-DV). The use of these techniques for long chain branch detection and the underlying theories have been well documented in the literature. See, for example, Zimm,
15 G.H. and Stockmayer, W.H., J. Chem. Phys., 17, 1301 (1949) and Rudin, A., Modern Methods of Polymer Characterization, John Wiley & Sons, New York (1991) pp. 103-112.

A. Willem deGroot and P. Steve Chum, both of The Dow Chemical Company, at the October 4, 1994 conference of the
20 Federation of Analytical Chemistry and Spectroscopy Society (FACSS) in St. Louis, Missouri, presented data demonstrating that GPC-DV is indeed a useful technique for quantifying the presence of long chain branches in substantially linear ethylene polymers. In particular, deGroot and Chum found
25 that the level of long chain branches in substantially linear ethylene homopolymer samples measured using the Zimm-Stockmayer equation correlated well with the level of long chain branches measured using ^{13}C NMR.

Further, deGroot and Chum found that the presence of
30 octene does not change the hydrodynamic volume of the polyethylene samples in solution and, as such, one can account for the molecular weight increase attributable to

octene short chain branches by knowing the mole percent octene in the sample. By deconvoluting the contribution to molecular weight increase attributable to 1-octene short chain branches, deGroot and Chum showed that GPC-DV may be
5 used to quantify the level of long chain branches in substantially linear ethylene/octene copolymers.

DeGroot and Chum also showed that a plot of $\text{Log}(I_2, \text{ melt index})$ as a function of $\text{Log}(\text{GPC Weight Average Molecular Weight})$ as determined by GPC-DV illustrates that the long
10 chain branching aspects (but not the extent of long branching) of substantially linear ethylene polymers are comparable to that of high pressure, highly branched low density polyethylene (LDPE) and are clearly distinct from ethylene polymers produced using Ziegler-type catalysts such
15 as titanium complexes and ordinary homogeneous catalysts such as hafnium and vanadium complexes.

For substantially linear ethylene polymers, the empirical effect of the presence of long chain branching is manifested as enhanced rheological properties which are
20 quantified and expressed in terms of gas extrusion rheometry (GER) results and/or melt flow, I_{10}/I_2 , increases.

The substantially linear ethylene polymers used in the present invention are a unique class of compounds that are further defined in U.S. Patent No. 5,272,236, application
25 number 07/776,130, filed October 15, 1991; U.S. Patent No. 5,278,272, application number 07/939,281, filed September 2, 1992; and U.S. Patent No. 5,665,800, application number 08/730,766, filed October 16, 1996.

Substantially linear ethylene polymers differ
30 significantly from the class of polymers conventionally known as homogeneously branched linear ethylene polymers described above and, for example, by Elston in U.S. Patent No.

3,645,992. As an important distinction, substantially linear ethylene polymers do not have a linear polymer backbone in the conventional sense of the term "linear" as is the case for homogeneously branched linear ethylene polymers.

5 Substantially linear ethylene polymers also differ significantly from the class of polymers known conventionally as heterogeneously branched traditional Ziegler polymerized linear ethylene interpolymers (for example, ultra low density polyethylene, linear low density polyethylene or high density
10 polyethylene made, for example, using the technique disclosed by Anderson et al. in U.S. Patent No. 4,076,698, in that substantially linear ethylene interpolymers are homogeneously branched polymers. Further, substantially linear ethylene
15 polymers also differ from the class of heterogeneously branched ethylene polymers in that substantially linear ethylene polymers are characterized as essentially lacking a measurable high density or crystalline polymer fraction as determined using a temperature rising elution fractionation technique.

20 The homogeneously branched substantially linear ethylene polymers for use in the present invention is characterized as having

- (a) melt flow ratio, $I_{10}/I_2 \geq 5.63$,
- (b) a molecular weight distribution, M_w/M_n , as
25 determined by gel permeation chromatography and defined by the equation:

$$(M_w/M_n) \leq (I_{10}/I_2) - 4.63,$$

- (c) a gas extrusion rheology such that the critical
30 shear rate at onset of surface melt fracture for the substantially linear ethylene polymer is at least 50 percent greater than the critical shear rate at the onset of surface melt fracture for a

linear ethylene polymer, wherein the substantially linear ethylene polymer and the linear ethylene polymer comprise the same comonomer or comonomers, the linear ethylene polymer has an I_2 and M_w/M_n within ten percent of the substantially linear ethylene polymer and wherein the respective critical shear rates of the substantially linear ethylene polymer and the linear ethylene polymer are measured at the same melt temperature using a gas extrusion rheometer,

(d) a single differential scanning calorimetry, DSC, melting peak between

-30° and 150°C, and

(e) a density less than or equal to 0.865 g/cm³.

Determination of the critical shear rate and critical shear stress in regards to melt fracture as well as other rheology properties such as "rheological processing index" (PI), is performed using a gas extrusion rheometer (GER). The gas extrusion rheometer is described by M. Shida, R.N. Shroff and L.V. Cancio in Polymer Engineering Science, Vol. 17, No. 11, p. 770 (1977) and in Rheometers for Molten Plastics by John Dealy, published by Van Nostrand Reinhold Co. (1982) on pp. 97-99.

The processing index (PI) is measured at a temperature of 190°C, at nitrogen pressure of 2500 psig (17.2 Mpa) using a 0.0296 inch (752 micrometers) diameter (preferably a 0.0143 inch diameter die for high flow polymers, for example, 50 - 100 I_2 melt index or greater), 20:1 L/D die having an entrance angle of 180°. The GER processing index is calculated in millipoise units from the following equation:

$$PI = 2.15 \times 10^6 \text{ dyne/cm}^2 / (1000 \times \text{shear rate}),$$

where: 2.15×10^6 dyne/cm² is the shear stress at 2500 psi (17.2 Mpa), and the shear rate is the shear rate at the wall as represented by the following equation:

$$32 Q' / (60 \text{ sec/min}) (0.745) (\text{Diameter} \times 2.54 \text{ cm/in})^3,$$

5 where:

Q' is the extrusion rate (gms/min),

0.745 is the melt density of polyethylene (gm/cm³),

and

Diameter is the orifice diameter of the capillary
10 (inches).

The PI is the apparent viscosity of a material measured at apparent shear stress of 2.15×10^6 dyne/cm².

For substantially linear ethylene polymers, the PI is less than or equal to 70 percent of that of a conventional
15 linear ethylene polymer having an I_2 , M_w/M_n and density each within ten percent of the substantially linear ethylene polymer.

An apparent shear stress vs. apparent shear rate plot is used to identify the melt fracture phenomena over a range of
20 nitrogen pressures from 5250 to 500 psig (36.2 to 3.4 Mpa) using the die or GER test apparatus previously described. According to Ramamurthy in Journal of Rheology, 30(2), 337-357, 1986, above a certain critical flow rate, the observed extrudate irregularities may be broadly classified
25 into two main types: surface melt fracture and gross melt fracture.

Surface melt fracture occurs under apparently steady flow conditions and ranges in detail from loss of specular gloss to the more severe form of "sharkskin". In this
30 disclosure, the onset of surface melt fracture is characterized at the beginning of losing extrudate gloss at

which the surface roughness of extrudate can only be detected by 40x magnification. The critical shear rate at onset of surface melt fracture for the substantially linear ethylene polymers is at least 50 percent greater than the critical
5 shear rate at the onset of surface melt fracture of a linear ethylene polymer having about the same I_2 and M_w/M_n .

Preferably, the critical shear stress at onset of surface melt fracture for the substantially linear ethylene polymers of the invention is greater than 2.8×10^6 dyne/cm².

10 Gross melt fracture occurs at unsteady flow conditions and ranges in detail from regular (alternating rough and smooth, helical, etc.) to random distortions. For commercial acceptability, (for example, in blown film products), surface defects should be minimal, if not absent. The critical shear
15 rate at onset of surface melt fracture (OSMF) and critical shear stress at onset of gross melt fracture (OGMF) will be used herein based on the changes of surface roughness and configurations of the extrudates extruded by a GER. For the substantially linear ethylene polymers used in the invention,
20 the critical shear stress at onset of gross melt fracture is preferably greater than 4×10^6 dyne/cm².

For the processing index determination and for the GER melt fracture determination, substantially linear ethylene polymers are tested without inorganic fillers and do not have
25 more than 20 ppm aluminum catalyst residue. Preferably, however, for the processing index and melt fracture tests, substantially linear ethylene polymers do contain antioxidants such as phenols, hindered phenols, phosphites or phosphonites, preferably a combination of a phenol or
30 hindered phenol and a phosphite or a phosphonite.

The molecular weight distributions of ethylene polymers are determined by gel permeation chromatography (GPC) on a

Waters 150C high temperature chromatographic unit equipped with a differential refractometer and three columns of mixed porosity. The columns are supplied by Polymer Laboratories and are commonly packed with pore sizes of 10^3 , 10^4 , 10^5 and 10^6\AA . The solvent is 1,2,4-trichlorobenzene, from which 0.3 percent by weight solutions of the samples are prepared for injection. The flow rate is 1.0 milliliters/minute, unit operating temperature is 140°C and the injection size is 100 microliters.

The molecular weight determination with respect to the polymer backbone is deduced by using narrow molecular weight distribution polystyrene standards (from Polymer Laboratories) in conjunction with their elution volumes. The equivalent polyethylene molecular weights are determined by using appropriate Mark-Houwink coefficients for polyethylene and polystyrene (as described by Williams and Ward in Journal of Polymer Science, Polymer Letters, Vol. 6, p. 621, 1968) to derive the following equation:

$$M_{\text{polyethylene}} = a * (M_{\text{polystyrene}})^b.$$

In this equation, $a = 0.4316$ and $b = 1.0$. Weight average molecular weight, M_w , is calculated in the usual manner according to the following formula: $M_j = (\sum w_i (M_i^j))^j$. Where w_i is the weight fraction of the molecules with molecular weight M_i eluting from the GPC column in fraction i and $j = 1$ when calculating M_w and $j = -1$ when calculating M_n .

For the at least one homogeneously branched ethylene polymer used in the present invention, the M_w/M_n is preferably less than 3.5, more preferably less than 3.0, most preferably less than 2.5, and especially in the range of from 1.5 to 2.5 and most especially in the range from 1.8 to 2.3.

Substantially linear ethylene polymers are known to have

excellent processability, despite having a relatively narrow molecular weight distribution (that is, the M_w/M_n ratio is typically less than 3.5). Surprisingly, unlike homogeneously and heterogeneously branched linear ethylene polymers, the melt flow ratio (I_{10}/I_2) of substantially linear ethylene polymers can be varied essentially independently of the molecular weight distribution, M_w/M_n . Accordingly, especially when good extrusion processability is desired, the preferred ethylene polymer for use in the present invention is a homogeneously branched substantially linear ethylene interpolymer.

Suitable constrained geometry catalysts for use manufacturing substantially linear ethylene polymers include constrained geometry catalysts as disclosed in U.S. application number 07/545,403, filed July 3, 1990; U.S. application number 07/758,654, filed September 12, 1991; U.S. Patent No. 5,132,380 (application number 07/758,654); U.S. Patent No. 5,064,802 (application number 07/547,728); U.S. Patent No. 5,470,993 (application number 08/241,523); U.S. Patent No. 5,453,410 (application number 08/108,693); U.S. Patent No. 5,374,696 (application number 08/08,003); U.S. Patent No. 5,532,394 (application number 08/295,768); U.S. Patent No. 5,494,874 (application number 08/294,469); and U.S. Patent No. 5,189,192 (application number 07/647,111).

Suitable catalyst complexes may also be prepared according to the teachings of WO 93/08199, and the patents issuing therefrom. Further, the monocyclopentadienyl transition metal olefin polymerization catalysts taught in U.S. Patent No. 5,026,798 are also believed to be suitable for use in preparing the polymers of the present invention, so long as the polymerization conditions substantially conform to those described in U.S. Patent No. 5,272,236; U.S.

Patent No. 5,278,272 and U.S. Patent No. 5,665,800, especially with strict attention to the requirement of continuous polymerization. Such polymerization methods are also described in PCT/US 92/08812 (filed October 15, 1992).

5 The foregoing catalysts may be further described as comprising a metal coordination complex comprising a metal of groups 3-10 or the Lanthanide series of the Periodic Table of the Elements and a delocalize β -bonded moiety substituted with a constrain-inducing moiety, said complex having a
10 constrained geometry about the metal atom such that the angle at the metal between the centroid of the delocalized, substituted pi-bonded moiety and the center of at least one remaining substituent is less than such angle in a similar complex containing a similar pi-bonded moiety lacking in such
15 constrain-inducing substituent, and provided further that for such complexes comprising more than one delocalized, substituted pi-bonded moiety, only one thereof for each metal atom of the complex is a cyclic, delocalized, substituted pi-bonded moiety. The catalyst further comprises an activating
20 cocatalyst.

Suitable cocatalysts for use herein include polymeric or oligomeric aluminoxanes, especially methyl aluminoxane, as well as inert, compatible, noncoordinating, ion forming compounds. So called modified methyl aluminoxane (MMAO) is
25 also suitable for use as a cocatalyst. One technique for preparing such modified aluminoxane is disclosed in U.S. Patent No. 5,041,584. Aluminoxanes can also be made as disclosed in U.S. Patent No. 5,218,071; U.S. Patent No. 5,086,024; U.S. Patent No. 5,041,585; U.S. Patent No.
30 5,041,583; U.S. Patent No. 5,015,749; U.S. Patent No. 4,960,878; and U.S. Patent No. 4,544,762.

Aluminoxanes, including modified methyl aluminoxanes, when used in the polymerization, are preferably used such that the catalyst residue remaining in the (finished) polymer is preferably in the range of from 0 to 20 ppm aluminum, especially from 0 to 10 ppm aluminum, and more preferably from 0 to 5 ppm aluminum. In order to measure the bulk polymer properties (for example, PI or melt fracture), aqueous HCl is used to extract the aluminoxane from the polymer. Preferred cocatalysts, however, are inert, noncoordinating, boron compounds such as those described in EP 520732.

Substantially linear ethylene are produced via a continuous (as opposed to a batch) controlled polymerization process using at least one reactor (for example, as disclosed in WO 93/07187, WO 93/07188, and WO 93/07189), but can also be produced using multiple reactors (for example, using a multiple reactor configuration as described in U.S. Patent No. 3,914,342) at a polymerization temperature and pressure sufficient to produce the interpolymers having the desired properties. The multiple reactors can be operated in series or in parallel, with at least one constrained geometry catalyst employed in at least one of the reactors.

Substantially linear ethylene polymers can be prepared via the continuous solution, slurry, or gas phase polymerization in the presence of a constrained geometry catalyst, such as the method disclosed in European Patent No. 416,815-A. The polymerization can generally be performed in any reactor system known in the art including, but not limited to, a tank reactor(s), a sphere reactor(s), a recycling loop reactor(s) or combinations thereof, any reactor or all reactors operated partially or completely adiabatically, nonadiabatically or a combination of both.

Preferably, a continuous loop-reactor solution polymerization process is used to manufacture the substantially linear ethylene polymer used in the present invention.

In general, the continuous polymerization required to manufacture substantially linear ethylene polymers may be accomplished at conditions well known in the prior art for Ziegler-Natta or Kaminsky-Sinn type polymerization reactions, that is, temperatures from 0 to 250°C and pressures from atmospheric to 1000 atmospheres (100 MPa). Suspension, solution, slurry, gas phase or other process conditions may be employed if desired.

A support may be employed in the polymerization, but preferably the catalysts are used in a homogeneous (that is, soluble) manner. It will, of course, be appreciated that the active catalyst system forms *in situ* if the catalyst and the cocatalyst components thereof are added directly to the polymerization process and a suitable solvent or diluent, including condensed monomer, is used in said polymerization process. It is, however, preferred to form the active catalyst in a separate step in a suitable solvent prior to adding the same to the polymerization mixture.

The substantially linear ethylene polymers used in the present invention are interpolymers of ethylene with at least one C₃-C₂₀ α -olefin and/or C₄-C₁₈ diolefin. Copolymers of ethylene and an α -olefin of C₃-C₂₀ carbon atoms are especially preferred. The term "interpolymer" as discussed above is used herein to indicate a copolymer, a terpolymer, or any other multiple monomer polymer, where at least one other comonomer is polymerized with ethylene or propylene to make the interpolymer.

Suitable unsaturated comonomers useful for polymerizing with ethylene include, for example, ethylenically unsaturated

monomers, conjugated or non-conjugated dienes, polyenes, etc. Examples of such comonomers include C₃-C₂₀ α -olefins such as propylene, isobutylene, 1-butene, 1-hexene, 1-pentene, 4-methyl-1-pentene, 1-heptene, 1-octene, 1-nonene, and 1-decene,. Preferred comonomers include propylene, 1-butene, 1-pentene, 1-hexene, 4-methyl-1-pentene and 1-octene, and 1-octene is especially preferred. Other suitable monomers include styrene, halo- or alkyl-substituted styrenes, vinylbenzocyclobutane, 1,4-hexadiene, 1,7-octadiene, and naphthenics (for example, cyclopentene, cyclohexene and cyclooctene).

The homogeneously branched ethylene interpolymer can be blended with other polymers. Suitable polymers for blending with the ethylene interpolymer are commercially available from a variety of suppliers and include, but are not limited, an ethylene polymer (for example, low density polyethylene, ultra or very low density polyethylene, medium density polyethylene, linear low density polyethylene, high density polyethylene, homogeneously branched linear ethylene polymer, substantially linear ethylene polymer, polystyrene, ethylene styrene interpolymer, ethylene vinyl acetate interpolymer, ethylene acrylic acid interpolymer, ethylene ethyl acetate interpolymer, ethylene methacrylic acid interpolymer, and ethylene methacrylic acid ionomer), polycarbonate, polystyrene, polypropylene (for example, homopolymer polypropylene, polypropylene copolymer, and random block polypropylene interpolymer), thermoplastic polyurethane, polyamide, polylactic acid interpolymer, thermoplastic block polymer (for example, styrene butadiene copolymer, styrene butadiene styrene triblock copolymer, and styrene ethylene-butylene styrene triblock copolymer), polyether block copolymer (for example, PEBAX), copolyester polymer,

polyester/polyether block polymers (for example, HYTREL),
ethylene carbon monoxide interpolymers (for example,
ethylene/carbon monoxide (ECO), copolymer, ethylene/acrylic
acid/carbon monoxide (EAACO) terpolymer, ethylene/methacrylic
5 acid/carbon monoxide (EMAACO) terpolymer, ethylene/vinyl
acetate/carbon monoxide (EVACO) terpolymer and styrene/carbon
monoxide (SCO)), polyethylene terephthalate (PET),
chlorinated polyethylene and mixtures thereof.

In one preferred embodiment, the homogeneously branched
10 ethylene interpolymers are blended with a polypropylene resin,
preferably an isotactic polypropylene resin such as Montell
Profax 6323 and Amoco 4018. However, generally suitable
polypropylene polymers for use in the invention, including
random block propylene-ethylene polymers, are available from
15 a number of manufacturers, such as, for example, Montell
Polyolefins, DSM, Amoco, Eastman, Fina and Exxon Chemical
Company. At Exxon, suitable polypropylene polymers are
supplied under the designations ESCORENE and ACHIEVE.

Suitable poly lactic acid (PLA) polymers for use in the
20 invention are well known in the literature (for example, see
D. M. Bigg et al., "Effect of Copolymer Ratio on the
Crystallinity and Properties of Polylactic Acid Copolymers",
ANTEC '96, pp. 2028-2039; WO 90/01521; EP 0 515203A; and EP 0
748846A2). Suitable poly lactic acid polymers are supplied
25 commercially by Cargill Dow under the designation EcoPLA.

Suitable thermoplastic polyurethane for use in the
invention are commercially available from The Dow Chemical
Company under the designation PELLATHANE.

Suitable polyolefin carbon monoxide interpolymers can be
30 manufactured using well known high pressure free-radical
polymerization methods. However, they may also be
manufactured using traditional Ziegler-Natta catalysis and

even with the use of so-called homogeneous catalyst systems such as those described and referenced herein above.

Suitable free-radical initiated high pressure carbonyl-containing ethylene polymers such as ethylene acrylic acid
5 interpolymers can be manufactured by any technique known in the art including the methods taught by Thomson and Waples in U.S. Patent No. 3,520,861 and by McKinney et al. in U.S. Nos. 4,988,781; 4,599,392; and 5,384,373.

Suitable ethylene vinyl acetate interpolymers for use in
10 the invention are commercially available from various suppliers, including Exxon Chemical Company and Du Pont Chemical Company.

Suitable ethylene/alkyl acrylate interpolymers are commercially available from various suppliers. Suitable
15 ethylene/acrylic acid interpolymers are commercially available from The Dow Chemical Company under the designation PRIMACOR. Suitable ethylene/methacrylic acid interpolymers are commercially available from Du Pont Chemical Company under the designation NUCREL.

20 Chlorinated polyethylene (CPE), especially chlorinated substantially linear ethylene polymers, can be prepared by chlorinating polyethylene in accordance with well known techniques. Preferably, chlorinated polyethylene comprises equal to or greater than 30 weight percent chlorine.

25 Suitable chlorinated polyethylenes for use in the invention are commercially supplied by The Dow Chemical Company under the designation TYRIN.

Suitable nitrogen-containing stabilizers for use in the present invention include, but are not limited to,
30 naphthylamines (for example, N-phenyl naphthylamines such as Naugard PAN supplied by Uniroyal); diphenylamine and derivatives thereof which are also referred to as secondary

aromatic amines (for example, 4, 4'-bis(α,α -dimethylbenzyl)-diphenylamine which is supplied by Uniroyal Chemical under the designation Naugard[®] 445); p-phenylenediamines (for example, Wingstay[®] 300 supplied by Goodyear); piperidines and
5 derivatives thereof (for example, polymeric N, N'-bis(2, 2, 6, 6-tetramethyl-4-piperidinyl)-1,6-hexanediamine with 2, 4, 6-trichloro-1, 3, 5-triazine and 2, 4, 4-trimethyl-1, 2-pentanamine which is supplied by Ciba-Geigy under the designation of Chimassorb[®] 944 as well as other substituted
10 piperidines such as Chimassorb[®] 119, Tinuvin[®] 622 and Tinuvin[®] 770, all three also supplied by Ciba-Geigy), and quinolines (for example, oxyquinolines and hydroquinolines such as polymeric 2, 2, 4-trimethyl-1, 2-dihydroquinoline which is supplied by Vanderbilt Company under the designation s
15 Agerite[®] D).

Suitable nitrogen-containing stabilizers also include the hybrid stabilizers such as aminophenols (for example, N, N'-hexamethylenebis-3-(3, 5-di-tert-butyl-4-hydroxyphenyl)-propionamide), acylaminophenols (which are also referred to
20 as 4-hydroxyanilides) and the various hybrid stabilizers described in U.S. Patent No. 5,122,593 which consist of a N-(substituted)-1-(piperazine-2-one alkyl) group at one end and a (3, 5-dialkyl-4-hydroxyphenyl)- α,α -disubstituted acetamine at the other end.

25 Other suitable nitrogen-containing stabilizers include carboxylic acid amides of aromatic mono and dicarboxylic acids and N-monosubstituted derivatives (for example, N, N'-diphenyloxamide and 2, 2'-oxamidobisethyl 3-(3, 5-di-tert-butyl-4-hydroxyphenyl) propionate which is supplied by
30 Uniroyal Chemical under the designation Naugard[®] XL-1); hydrazides of aliphatic and aromatic mono- and dicarboxylic acids and N-acylated derivatives thereof; bis-acylated

hydrazine derivatives; melamine; benzotriazoles, hydrazones;
acylated derivatives of hydrazino-triazines; polyhydrazides;
salicylaethylenediimines; salicylaloximes; derivatives of
ethylenediamino tetraacetic acid; and aminotriazoles and
5 acylated derivatives thereof.

Preferred nitrogen-containing stabilizers for use in the
present invention are diphenylamines, substituted piperidines
and hydroquinolines. Further, the at least one nitrogen-
containing stabilizer can be employed alone or in combination
10 with another stabilizer and antioxidant such as, for example,
but not limited to, other nitrogen-containing stabilizer as
well as a hindered phenol (for example, 2, 6-di-tert-butyl-4-
methylphenol which is supplied by Koppers Chemical under the
designation BHT[®] and tetrakis(methylene 3-(3, 5-di-tert-butyl-
15 4-hydroxyphenyl) propionate) methane which is supplied by
Ciba-Geigy under the designation Irganox[®] 1010); thioester
(for example, dilauryl thiodipropionate which is supplied by
Evans under the designation Evanstab[®] 12); phosphite (for
example, Irgafos[®] 168 supplied by Ciba-Geigy Corp. and
20 tri(nonylphenyl) phosphite which is supplied by Uniroyal
Chemical under the designation Naugard[®] P); diphosphite (for
example, distearyl pentaerthritol diphosphite which is
supplied by Borg-Warner under the designation Weston[®] 618);
polymeric phosphite (for example, Wyttox[®] 345-S(1) supplied by
25 Olin); phosphited phenol and bisphenol (for example, Wyttox[®]
604 supplied by Olin); and diphosponite (for example,
tetrakis(2, 4-di-tert-butylphenyl) 4, 4'-biphenylene
diphosponite which is supplied by Sandox under the
designation Sandostab[®] P-EPQ).

30 The at least one nitrogen-containing stabilizer is add
to the homogeneously branched ethylene polymer in a melt
compounding step, preferably by the use of an additive

concentrate, prior to fabrication and shaping process steps. The at least one nitrogen-containing stabilizer can be added to the interpolymer at any effective concentration. However, preferably, the concentration of the least one nitrogen-
5 containing stabilizer will for in the range of from 0.05 to 0.5 weight percent (based on the total weight of the stabilizer and interpolymer), more preferably in the range from 0.075 to 0.3 weight percent (based on the total weight of the stabilizer and the interpolymer) and most preferably
10 in the range of from 0.1 to 0.25 weight percent (based on the total weight of the stabilizer and the interpolymer).

In-process additives, for example, calcium stearate, water, and fluoropolymers, may also be used for purposes such as for the deactivation of residual catalyst or improved
15 processability or both. Colorants, coupling agents and fire retardants may also be include as longer as their incorporation does not disturb the desirable characteristics of the inventive article, interpolymer or method.

The homogeneously branched ethylene interpolymer can also be filled or unfilled. If filled, then the amount of
20 filler present should not exceed an amount that would adversely affect elevated temperature elasticity and/or the wash and dryability of the inventive article. Typically, the amount of filler present is between 20 and 80, preferably
25 between 50 and 70, weight percent (wt percent) based on the total weight of the interpolymer. Representative fillers include kaolin clay, magnesium hydroxide, silica, calcium carbonate. In a preferred embodiment, in which a filler is present, the filler is coated with a material that will
30 prevent or retard any tendency that the filler might otherwise have to interfere with desirable irradiation and/or crosslinking reactions and effects. Stearic acid is an

illustrative example of such a protective filler coating, although other compound may be employed as protective filler coatings or treatments.

The improved elastic homogeneously branched ethylene
5 interpolymer and elastic article of the invention have utility in a variety of applications. Suitable applications include, for example, but are not limited to, disposable personal hygiene products (for example, training pants, diapers, absorbent underpants, incontinence products, and
10 feminine hygiene items); disposable and durable garments (for example, elastic components in industrial apparel, coveralls, head coverings, underpants, pants, shirts, gloves, and socks); infection control/clean room products (for example, surgical gowns and drapes, face masks, head coverings,
15 surgical caps and hood, shoe coverings, boot slippers, wound dressings, bandages, sterilization wraps, wipers, lab coats, coverall, pants, aprons, jackets, and bedding items and sheets) and sports apparel.

Various homofil fibers can be made from the elastic
20 ethylene interpolymer of the present invention, including staple fibers, spunbond fibers or melt blown fibers (using, for example, systems as disclosed in U.S. Patent No. 4,340,563 (Appel et al.), U.S. Patent No. 4,663,220 (Wisneski et al.), U.S. Patent No. 4,668,566 (Braun), or U.S. Patent No.
25 4,322,027 (Reba)), and gel spun fibers (for example, the system disclosed in U.S. Patent No. 4,413,110 (Kavesh et al.))). Staple fibers can be melt spun (that is, they can be extruded into the final fiber diameter directly without additional drawing), or they can be melt spun into a higher
30 diameter and subsequently hot or cold drawn to the desired diameter using conventional fiber drawing techniques.

Elastic staple fibers of the present invention herein can also be used as bonding fibers, especially where the inventive elastic fibers have a lower melting point than the surrounding matrix fibers. In a bonding fiber application, the bonding
5 fiber is typically blended with other matrix fibers and the entire structure is subjected to heat, where the bonding fiber melts and bonds the surrounding matrix fiber. Typical matrix fibers which benefit from use of the inventive elastic fibers disclosed herein include, but are not limited to,
10 poly(ethylene terephthalate) fibers, cotton fibers, nylon fibers, polypropylene fibers, heterogeneously branched polyethylene fibers, homogeneously branched ethylene polymer fibers, and linear polyethylene homopolymer fibers and combinations thereof. The diameter of the matrix fiber can
15 vary depending upon the end use application.

Bicomponent fibers can also be made from the inventive elastic interpolymer described herein: Such bicomponent fibers have the elastic homogeneously branched ethylene interpolymer of the present invention in at least one portion
20 of the fiber. For example, in a sheath/core bicomponent fiber (that is, one in which the sheath concentrically surrounds the core), the stable, elastic homogeneously branched ethylene interpolymer can be in either the sheath or the core. Different elastic homogeneously branched ethylene
25 interpolymers of the present invention can also be used independently as the sheath and the core in the same fiber, preferably where both components are elastic and especially where the sheath component has a lower melting point than the core component. Other types of bicomponent fibers are within
30 the scope of the invention as well, and include such structures as side-by-side conjugated fibers (for example, fibers having separate regions of polymers, wherein the

elastic homogeneously branched ethylene interpolymer of the present invention comprises at least a portion of the fiber's surface).

The shape of the fiber is not limited. For example,
5 typical fiber has a circular cross-sectional shape, but sometimes fibers have different shapes, such as a trilobal shape, or a flat (that is, "ribbon" like) shape. The elastic fiber disclosed herein is not limited by the shape of the fiber.

10 Fiber diameter can be measured and reported in a variety of fashions. Generally, fiber diameter is measured in denier per filament. Denier is a textile term which is defined as the grams of the fiber per 9000 meters of that fiber's length. Monofilament generally refers to an extruded strand having a
15 denier per filament greater than 15, usually greater than 30. Fine denier fiber generally refers to fiber having a denier of 15 or less. Microdenier (aka microfiber) generally refers to fiber having a diameter not greater than 100 micrometers. For the inventive elastic fibers disclosed herein, the diameter
20 can be widely varied, with little impact upon the fiber's elasticity. However, the fiber denier can be adjusted to suit the capabilities of the finished article and as such, would preferably be: from 0.5 to 30 denier/filament for melt blown; from 1 to 30 denier/filament for spunbond; and from 1 to
25 20,000 denier/filament for continuous wound filament.

Fabrics made from the inventive stable, elastic fibers disclosed herein include both woven and nonwoven fabrics. Nonwoven fabrics can be made variously, including spunlaced (or hydrodynamically entangled) fabrics as disclosed in U.S.
30 Patent No. 3,485,706 (Evans) and U.S. Patent No. 4,939,016 (Radwanski et al.); by carding and thermally bonding staple fibers; by spunbonding continuous fibers in one continuous

operation; or by melt blowing fibers into fabric and subsequently calendering or thermally bonding the resultant web. These various nonwoven fabric manufacturing techniques are well known to those skilled in the art and the disclosure
5 is not limited to any particular method. Other structures made from such fibers are also included within the scope of the invention, including for example, blends of these novel stable, elastic fibers with other fibers (for example, poly(ethylene terephthalate) (PET) or cotton).

10 Fabricated articles which can be made using the inventive stable, elastic fibers and fabrics disclosed herein include stable, elastic composite articles (for example, diapers and undergarments) that have elastic portions. For example, elastic portions are typically constructed into
15 diaper and undergarment waist band portions to prevent the diaper or undergarment from falling and into leg band portions to prevent leakage (as shown, for example, in U.S. Patent No. 4,381,781 (Sciaraffa)). Often, the elastic portions promote better form fitting and/or fastening systems
20 for a good combination of comfort and reliability.

The inventive stable elastic fibers and fabrics disclosed herein can also produce structures which combine elasticity with breathability. For example, the inventive elastic fibers, fabrics and/or films of the present
25 invention may be incorporated into the structures disclosed in U.S. provisional patent application 60/083,784, filed May 1, 1998 in name Maugans et al.

The inventive stable elastic fibers and fabrics disclosed herein can also be used in various structures as
30 described in U.S. Patent No. 2,957,512 (Wade). For example, layer 50 of the structure described in USP '512 (that is, the elastic component) can be replaced with the inventive stable

elastic fibers and fabrics, especially where flat, pleated, creped, crimped, etc., nonelastic materials are made into elastic structures. Attachment of the inventive stable elastic fibers and/or fabric disclosed herein to nonelastic
5 fibers, fabrics or other structures can be done by melt bonding or with adhesives. Gathered or shirred elastic structures can be produced from the inventive stable elastic fibers and/or fabrics disclosed herein and nonelastic components by pleating the non-elastic component (as
10 described in USP '512) prior to attachment, pre-stretching the elastic component prior to attachment, or heat shrinking the elastic component after attachment.

The inventive stable elastic fibers described herein also can be used in a spunlaced (or hydrodynamically
15 entangled) process to make novel structures. For example, U.S. Patent No. 4,801,482 (Goggans) discloses an elastic sheet (12) which can now be made with the novel elastic fibers/fabric described herein.

Continuous stable elastic filaments as described herein
20 could also be used in woven applications where high resilience is desired.

The inventive stable elastic fibers and fabrics disclosed herein with adjust in the interpolymer melt index and/or degree of crosslinking or extent or radiation also
25 have adjustable tenacity and retractive force. Such capabilities and characteristics enable extensive design flexibility, for example, to provide for variable retractive force in the same garment, if needed, as described for example in U.S. Patent No. 5,196,000 (Clear et al.).

30 U.S. Patent No. 5,037,416 (Allen et al.) describes the advantages of a form fitting top sheet by using elastic ribbons (see member 19 of USP' 416). The inventive stable

elastic fibers could serve the function of member 19 of USP '416, or could be used in fabric form to provide the desired elasticity.

Composites that utilize very high molecular weight
5 linear polyethylene or copolymer polyethylene also benefit from the inventive stable elastic fibers disclosed herein. For example, the inventive elastic fibers have a low melting point (with the melting point of the polymer essentially linearly related to the polymer density), such that in a
10 blend of inventive stable elastic fibers disclosed herein and very high molecular weight polyethylene fibers (for example, SpectraTM fibers made by Allied Chemical) as described in U.S. Patent No. 4,584,347 (Harpell et al.), the lower melting elastic fibers bond the high molecular weight polyethylene
15 fibers without melting the high molecular weight fibers, thus preserving the high strength and integrity of the high molecular weight fiber.

In U.S. Patent No. 4,981,747 (Morman), the inventive stable elastic fibers and/or fabrics disclosed herein can be
20 substituted for elastic sheet 122, which forms a composite elastic material including a reversibly necked material.

The inventive stable elastic fibers disclosed herein can also be a melt blown elastic component, as described in reference 6 of the drawings of U.S. Patent No. 4,879,170
25 (Radwanski). USP '170 generally describes elastic co-form material and manufacturing processes.

Elastic panels can also be made from the inventive stable elastic fibers and fabrics disclosed herein, and can be used, for example, as members 18, 20, 14, and/or 26 of
30 U.S. Patent No. 4,940,464 (Van Gompel). The inventive stable elastic fibers and fabrics described herein can also be used

as elastic components of composite side panels (for example, layer 86 of USP '464).

The stable elastic homogeneously branched ethylene interpolymer can also be shaped or fabricated into stable elastic films, coatings, sheets, strips, straps, and tapes, ribbons. The elastic film, coating and sheet of the present invention may be fabricated by any method known in the art, including blown bubble processes (for example, simple bubble as well as biaxial orientation techniques such trapped bubble, double bubble and tenter framing), cast extrusion, injection molding processes, thermoforming processes, extrusion coating processes, profile extrusion, and sheet extrusion processes. Simple blown bubble film processes are described, for example, in The Encyclopedia of Chemical Technology, Kirk-Othmer, Third Edition, John Wiley & Sons, New York, 1981, Vol. 16, pp. 416-417 and Vol. 18, pp. 191-192. The cast extrusion method is described, for example, in Modern Plastics Mid-October 1989 Encyclopedia Issue, Volume 66, Number 11, pages 256 to 257. Injection molding, thermoforming, extrusion coating, profile extrusion, and sheet extrusion processes are described, for example, in Plastics Materials and Processes, Seymour S. Schwartz and Sidney H. Goodman, Van Nostrand Reinhold Company, New York, 1982, pp. 527-563, pp. 632-647, and pp. 596-602.

The stable elastic strips, tapes and ribbons of the present invention can be prepared by any known method, including the direct extrusion processing or by post-extrusion slitting, cutting or stamping techniques. Profile extrusion is an example of a primary extrusion process that is particularly suited to the preparation of tapes, strips and bands, ribbons.

The stable elastic materials of the present invention can also be rendered pervious or "breathable" by any method well known in the art including by apperturing, slitting, microperforating, mixing with fibers or foams and combinations thereof. Examples of such methods include, U.S. Patent No. 3,156,242 by Crowe, Jr., U.S. Patent No. 3,881,489 by Hartwell, U.S. Patent No. 3,989,867 by Sisson and U.S. Patent No. 5,085,654 by Buell.

Fabricated articles which can be made using the inventive stable elastic articles disclosed herein include composite fabric articles (for example, disposable incontinence garments and diapers) that are comprised of one or more elastic component or portion. The inventive stable elastic articles disclosed herein can also produce fabric composite structures which combine elasticity with breathability by utilizing a technique that renders the elastic material pervious or "breathable" such as suggested by Lippert et al. in U.S. Patent No. 4,861,652 and indicated above.

The inventive stable elastic articles disclosed herein can also be used in various structures as described in U.S. Patent No. 2,957,512 (Wade). For example, layer 50 of the structure described in USP '512 (that is, the elastic component) can be replaced with the novel stable elastic materials, especially where flat, pleated, and creped nonelastic materials are made into elastic or semi-elastic structures. Attachment of the novel stable elastic materials to nonelastic or less-elastic materials can be done with heat bonding or with adhesives. Gathered or shirred elastic composite materials can be produced from the new stable elastic material described herein and nonelastic components by pleating the non-elastic component (as described in USP

'512) prior to attachment, prestretching the elastic component prior to attachment, or heat shrinking the elastic component after attachment.

The recovery after heat shrinking can be further enhanced by effectuating a high degree of orientation into the inventive stable elastic articles during fabrication. Significant orientation can be accomplished by the utilization of various known techniques such as high blow-up blown film fabrication, tenter framing of cast films and "double bubble" or "trapped bubble" blown film fabrication.

The inventive stable elastic articles described herein can also be used to make other novel structures. For example, U.S. Patent No. 4,801,482 (Goggans) discloses an elastic sheet (12) which can now be made with the inventive stable elastic articles described herein.

The inventive stable elastic articles described herein can also be used to make breathable portion or breathable elastic composite materials. For example, U.S. Patent No. 5,085,654 (Buell) discloses a leg band (15) with a breathable portion 45, a breathable topsheet (26), a breathable backsheet (25), elastic elements (31 and 64), a breathable element (54), and a breathable sub-element (96) all or any combination of which can now be made with the inventive stable elastic articles disclosed herein in either pervious or impervious forms.

U.S. Patent No. 5,037,416 (Allen et al.) describes the advantages of a form fitting top sheet by using elastic ribbons (member 12) and an elastic backsheet (member 16). Pervious stable inventive elastic articles described herein could serve the function of member 12 and impervious elastics materials of this invention could function as member 16, or

disclosed elastic materials could be used in an elastic composite fabric form.

In U.S. Patent No. 4,981,747 (Morman), the inventive stable elastic articles disclosed herein can be substituted
5 for elastic sheets 12, 122 and 232 to construct an elastic composite material which includes a reversibly necked material.

Elastic panels, elements, or portions can also be made from the inventive stable elastic articles disclosed herein,
10 and can be used, for example, as members 18, 20, 24, and/or 26 of U.S. Patent No. 4,940,464 (Van Gompel). The inventive stable elastic articles described herein can also be used, for example, as elastic composite side panels (for example, layer) or as elastic ribbons 42 and/or 44.

15 The following examples are provided to further illustrate and illuminate the present invention but is not intended to limit the invention to the specific embodiments set forth.

20 EXAMPLES

In an evaluation to determine the elastic performance of various ethylene polymers in response to irradiation or
25 crosslinking, five different ethylene interpolymers were subjected to varying degrees of electron beam radiation and their elastic properties as 2 mil (102 mm) cast films were measured at room temperature. The polymer densities and the melt indexes of the ethylene polymers are shown in Table 1.
30 All of the polymers were homogeneously branched ethylene/1-octene interpolymers supplied commercially by Dupont Dow Elastomers, Ltd., manufactured using a constrained geometry

catalyst system and contained 2000 ppm Irganox 1010 thermal stabilizer. However, DDE 8190 also contained via blending 4-5 weight percent of isotactic polypropylene. The densities for the various polymers were determined in accordance with
5 ASTM D-792 and the melt indexes were determined in accordance with ASTM D-1238 Condition 190°C/2.16 kilograms.

Table 1

Polymer	Density (g/cm ³)	Melt Index (g/10 minutes)	Designation
A	0.863	0.5	ENGAGE EG 8180
B	0.859	1.0	DDE 8190
C	0.870	1.0	ENGAGE EG 8100
D	0.870	5.0	ENGAGE EG 8200
E	0.870	10	XU-58380.00

10

2 mil (102 mm) cast films of each polymer listed in Table 1 were fabricated using conventional cast film extrusion equipment at melt temperatures of 430°-500°F (221°-260°C). After film fabrication, the cast films were electron
15 beam radiated at various dosage using equipment similar to that described in U.S. Patent No. 5,324, 576. Except as otherwise indicated, the elastic properties (stress-strain data) for the various films were determined using an Instron tensiometer set at 10 inch/minute (25 cm/min.).

20

For the permanent set determinations at 23°C, the gage length was 2 inches (5.1 cm) and the crosshead speed was 20 inches/minute (51 cm/min.). The test consisted of pulling the film sample to 200 percent strain (elongation) and holding it for 30 seconds, then taking the sample to 0
25 percent strain (elongation) and holding it at 0 percent strain for 60 seconds, and then pulling the sample to determine the point where the load initially rises above zero. The percent permanent set was taken as the percent

strain at which the load rose above zero. The test was a one cycle test ran in duplicate.

For percent stress or load relaxation determinations at 23°C, the gage length was 2 inches (5.1 cm) and the crosshead speed was 20 inches/minute (51 m/min.). These tests consisted of pulling film samples to 200 percent strain (elongation) and holding them there at 200 percent strain for 30 seconds. The stress initially at 200 percent strain was taken as the maximum stress and the stress after the 30 second holding period was taken as the minimum stress. The percent stress or load relaxation determinations were run in duplicate and was calculated from the following equation:

$$\frac{\text{maximum stress} - \text{minimum stress}}{\text{maximum stress}} \times 100.$$

Table 2 reports the elastic property (stress-strain) data as well as the permanent set and stress relaxation data for the various film samples.

Data in Table 2 were plotted and are shown in FIG. 1 and 2. FIG. 1 indicates that electron beam radiation up to 8-12 megarads has no substantial affect on the percent stress relaxation performance of the various polymers. Conversely, FIG. 2 shows that irradiation has a dramatic affect on the percent permanent set performance of the ethylene polymers. However, FIG. 2 (like FIG. 1 and the results shown in WO 95/29197) shows no particular distinction between the various polymers as polymer density dominated the percent permanent set response and radiation affected the various polymer equally.

In another evaluation, 2 mil (102 mm) cast films of Resin A and Resin D were subjected to varying dosages of electron beam radiation and evaluated to determine their respective percent stress or load relaxation performances at 38°C. These tests were performed as described above, except the temperature was 38°C instead of 23°C and the samples were held at 200 percent strain for 1 hour instead for 30 seconds. Table 3 shows the results for this evaluation and FIG. 3 plots the results using the average of duplicative samples as well as a four-datapoint average for Resin D at 5 and 8 megarads of electron beam radiation.

Table 3

E-Beam megarads	Maximum Stress, psi	Minimum Stress, psi	Percent Stress Relaxation
Resin D (Comp. Ex. 4)			
0	192	127	33.9
0	181	125	30.9
5	235	112	52.3
5	230	108	53.0
8	231	132	42.9
8	245	136	44.5
12	250	120	52.0
12	227	115	49.3
Resin A (Inv. Ex. 1)			
0	126	55	56.3
0	120	53	55.8
3	120	57	52.5
3	134	59	56.0
5	134	64	52.2
5	142	69	51.4
8	137	70	48.9
8	145	78	46.2

The data in Table 3 and FIG. 3 show, surprisingly, that irradiation can substantially reduce (that is, improve) the elevated temperature percent stress relaxation performance of ethylene interpolymers having densities less than 0.87 g/cm^3 and conversely, show irradiation has no affect or increases the elevated temperature stress relaxation performance of ethylene interpolymers characterized by higher densities. The data in Table 3 also show that the minimum stress of ethylene interpolymers characterized as having densities less than 0.87 g/cm^3 desirably increases at higher dosage level. Extrapolation of FIG. 3 indicates that at an electron beam radiation dosage level of 20 megarads, such interpolymers will exhibit a percent stress relaxation at 38°C of less than 20.

In another investigation, the effectiveness of various stabilizers was evaluated. In this investigation, 2 wt. percent stabilizer concentrates were prepared by first separately tumble dry blending a thioester stabilizer (that is, Evanstab 12), a diphenylamine stabilizer (Naugard 445), a substituted piperidine stabilizer (that is, Chimassorb 944) and a hydroquinoline stabilizer (that is, Agerite D) with a homogeneously branched ethylene interpolpolymer (ENGAGE 8150 which had a target 0.78 g/cm^3 density and 0.5 g/10 minutes I_2 melt index). The dry blends were then melt extruded in a Berlyne extruder having a 20:1 L/D and equipped with a 1-inch (2.5 cm) diameter screw. The extrusion melt temperature was maintained at 400°F (204°C), the various melts were pelletized and permitted to cool to ambient temperature.

Also, a 10 wt. percent concentrate of Irganox 1010 (a hindered phenolic stabilizer) was prepared using the same homogeneously branched ethylene interpolpolymer as above. However, the Irganox 1010 concentrate was prepared a Haake

compounder set at a melt temperature of 400°F (204°C) and a mixing residence time of 5 minutes. The Haake melt blend was removed from the mixing bowl after the 5 minute mixing residence time, permitted to cool to ambient temperature and then chopped into small granules.

The various stabilizer concentrates were tumble dry blended with two different homogeneously branched ethylene interpolymers, one nominally stabilized and the other non-stabilized, and both having a peak melting point less than 70°C as determined using differential scanning calorimetry, to prepared various samples. The samples were then separately melt spun into fibers.

The fibers were separately melt spun on fiber extrusion equipment consisting of an extruder, gear pump and spinneret. The extruder was set to provide a melt temperature of 236°C. Each polymer melt stream was feed to the gear pump which pressurized the melt and passed it through a 200 mesh pack followed by a 34-hole spinneret die. The spinneret had a 4:1 L/D and the holes had a diameter of 800 microns. The resin output from the spinneret was controlled at 0.78 gram/hole. The fibers were quenched with a room temperature air high-velocity blower and collected as free fall fiber samples. The resulting fibers an average diameter of 800 microns. All stabilizers permitted the preparation of good quality fiber as there were no process upset or fiber surface defects associated with any stabilizer.

The various fiber samples were then irradiated using electron-beam radiation at 20 megarads. The irradiated fibers were then evaluated to determine their ability to resistance ordinary washing and long-term oven aging. To determine stability or resistance, a fiber sample was stretched to 200 percent strain (elongation) five times using

an Instron tensiometer and then placed in a 160°F (71°C) wash solution. The wash solution consisted of 100 ppm of copper chloride and 0.5 weight percent Tide™ detergent regular household formula as supplied by Proctor and Gamble) and distilled water. The wash solution consisted of 275 milliliters contained in a 500 ml wide-mouth beaker. The 160°F (71°C) wash temperature was maintained by the use of a hot plate equipped with a Variac and a thermo-watch device. Agitation of the wash solution was accomplished using a magnetic stirrer wherein the hot-plate provided the counter magnetic field currents to effectuate rotation of the magnetic stirrer. All fiber samples were placed in the wash solution at once (together) and were vigorously agitated for 30 minutes. After the 30 minute exposure to the wash solution, the fiber samples were carefully removed using tweezers and placed on paper towels to absorb excess wash solution. The washed fibers were then sparsely placed on Mylar™ film (polyester A type film available from The Pilcher Hamilton Company) and placed in a circulating air oven. The oven was set to 133°C for 10 hours. After the 10-hour oven aging, the fiber samples were carefully (that is, avoiding excessive handling and direct handling) removed from the oven and examined for visual indications of loss of integrity (for example, melting and flowing or adhesion to the Mylar film or both). Table 4 sets forth the description of the various samples, provides descriptions of the samples following the wash and oven aging exposures and also provides a rank ordering of the resistance of the various samples to the wash/oven exposure. The results in Table 4 indicate that nitrogen-containing stabilizers such as Naugard 445, Chimassorb 944 and Agerite D are more effective than thioester or phenol stabilizer in stabilizing the elastic

fibers against loss of integrity due to washing and oven aging. In particular, Table 4 shows that stabilizers such as Agerite D can provide outstanding protection such that there is absolutely no melting of the fiber or adhesion to Mylar film.

Although the test described above can suitably and adequately distinguish the merits of the present invention, improved quantification can be conveniently accomplished by measuring fiber diameter (at the widest point) before the exposures and comparing those measurements to diameter measurements (at the widest point) taken after the exposures. Any measurable different in the diameters can be taken as a loss of integrity due to melting and flowing. However, a practitioner will recognize that indications of flowing-out or swelling are indicative of more substantial changes in integrity than melting and that adhesion to Mylar film generally reflects a less substantial change than melting.

Practitioners will also recognize that indication of melting must be made immediately when the sample is removed from the oven and that various aids can be used to help determine whether a polymer material is molten or not. Such aids may include, for example, a pin-point probe, a microscope, or Polaroid™ lenses.

	Resin	Additive (Target ppm)	Fiber Integrity Observations *	Rank Order
Comp. Ex. 6	F	None	melting; adhesion when upside down	8
Comp. Ex. 7	G	Evanstab 12 (1000)	melting; adhesion to 45- 90°	7
Comp. Ex. 8	G	None	flowing; melting; adhesion to 45- 90°	6
Comp. Ex. 9	G	Irganox 1010 (2000)	flowing; melting; adhesion to 30- 45°	5
Comp. Ex. 10	F	Irganox 1010 (2000)	adhesion to 45- 90°	4
Inv. Ex. 11	G	Naugard 445 (1000)	adhesion to <30°	3
Inv. Ex. 12	G	Chimassorb 944 (1000)	adhesion to <30°	2
Inv. Ex. 13	G	Agerite D (1000)	no adhesion; no melting; no flowing; no swelling	1

Resin F is an ethylene-octene copolymer having a target density of 0.870 g/cm³ and I₂ melt index of 5 g/10 minutes.

Resin G is an ethylene-octene copolymer having a target density of 0.870 g/cm³ and I₂ melt index of 0.5 g/10 minutes and containing 800 ppm Sandostab P-EPQ and 500 ppm Irganox 1076, Ciba-Geigy trademark for octadecyl 3-(3, 5-di-tert-butyl-4-hydroxyphenyl) propionate which is a hindered phenol stabilizer.

*The degree notation as to adhesion refers to the angle that the Mylar film had to be deflected to unstick the given sample where 0° was the flat horizontal

surface and 90° was approximately perpendicular to the flat horizontal surface, except Inv. Ex. 13 moved freely with the slightest handling of the film.

We Claim:

1. A method of making a shaped curable or crosslinkable article comprising at least one homogeneously branched ethylene interpolymer, which comprises ethylene
5 interpolymerized with at least one other monomer and characterized as having (before being shaped, grafted, cured, irradiated, or crosslinked) a polymer density of less than 0.90 g/cm³ at 23°C, and at least one nitrogen-containing stabilizer.
10
2. A method of making a shaped and cured, irradiated or crosslinked article comprising at least one homogeneously branched ethylene interpolymer, which comprises ethylene
15 interpolymerized with at least one other monomer and characterized as having (before being shaped, grafted, cured, irradiated, or crosslinked) a polymer density of less than 0.90 g/cm³ at 23°C, and at least one nitrogen-containing stabilizer.
- 20 3. A method of making an elastic article comprising the steps of:
 - (a) providing at least one homogeneously branched ethylene interpolymer having a density of less than or equal to 0.865 g/cm³ at 23°C having at least 0.05 weight
25 percent of at least one nitrogen-containing stabilizer therein,
 - (b) fabricating or shaping the article from the interpolymer, and
 - (c) after the fabrication or shaping, subjecting
30 the article to heat and/or ionizing radiation, wherein the article is characterized as having:

- (i) a percent permanent set of less than 60 at 23°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using an Instron tensiometer after being shaped and cured, irradiated or crosslinked,
- 5 (ii) a percent stress relaxation of less than or equal 25 at 23°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using a Instron tensiometer after being shaped and cured, irradiated or crosslinked, and
- 10 (iii) a percent stress relaxation of less than or equal 55 at 38°C and 200 percent strain when measured at a 2 mil (102 mm) thickness using an Instron tensiometer after.
- 15 4. The methods of Claims 1, 2 and 3 wherein the method further comprises incorporating at least one pro-rad additive into the interpolymer.
- 20 5. A method of making a curable elastic article comprising the steps of:
- (a) providing at least one homogeneously branched ethylene interpolymer characterized as having a density at 23°C less than 0.90 g/cm³ and comprising at least 0.1 weight percent of at least one nitrogen-containing stabilizer
- 25 incorporated therein,
- (b) preparing a melt of the stabilized interpolymer of (a);
- (c) mixing into the melt of (b) from 0.5 to 5 phr of a silane crosslinker (parts of silane crosslinker per hundred
- 30 parts interpolymer) while the crosslinker is at an ambient temperature between 0 and 30°C; and

(d) subjecting the melt mixture of (c) to ionizing energy or contacting the melt mixture of (c) with at least one free radical initiator to graft at least 50 weight percent, based on the total weight of the crosslinker and the
5 interpolymers, of the silane crosslinker to the stabilized interpolymers.

6. The method of Claim 1, wherein the at least one homogeneously branched ethylene interpolymers is a
10 substantially linear ethylene interpolymers characterized as having

(a) melt flow ratio, $I_{10}/I_2 \geq 5.63$,

(b) a molecular weight distribution, M_w/M_n , as determined by gel permeation chromatography and
15 defined by the equation:

$$(M_w/M_n) \leq (I_{10}/I_2) - 4.63,$$

(c) a gas extrusion rheology such that the critical shear rate at onset of surface melt fracture for the substantially linear ethylene polymer is at
20 least 50 percent greater than the critical shear rate at the onset of surface melt fracture for a linear ethylene polymer, wherein the substantially linear ethylene polymer and the linear ethylene polymer comprise the same comonomer or comonomers,
25 the linear ethylene polymer has an I_2 and M_w/M_n within ten percent of the substantially linear ethylene polymer and wherein the respective critical shear rates of the substantially linear ethylene polymer and the linear ethylene polymer
30 are measured at the same melt temperature using a gas extrusion rheometer, and

- (d) a single differential scanning calorimetry, DSC, melting peak between -30° and 150°C.

5 7. The method of Claim 3, wherein the ionizing radiation is provided by electron beam irradiation.

8. The method of Claim 1, wherein at least one nitrogen-containing stabilizer is selected from the group
10 consisting of a hydroquinoline, diphenylamine and substituted piperidine.

9. The method of Claim 3, wherein the article is fabricated using a technique selected from the group
15 consisting of fiber melt spinning, fiber melt blowing, spunbonding, spunlacing, carding, film blowing, cast film, injection molding, pultrusion, thermoforming, stamping, forging, blow molding, sheet extrusion, solvent casting, solvent coating, thermal lamination, calendering, roll
20 milling, reaction injection molding, extrusion coating, dispersion coating, and rotomolding.

10. The method of Claim 3 wherein the article is permitted to cool or quenched to ambient temperatures between
25 0 and 30°C before the application of additional heating or ionizing radiation.

11. The method of Claim 1, wherein the homogeneously branched ethylene interpolpolymer is a homogeneously branched
30 linear ethylene polymer.

12. The method of Claim 10, wherein the homogeneously branched linear ethylene interpolpolymer is characterized as having a single differential scanning calorimetry, DSC, melting peak between -30° and 150°C, and

5

13. The method of Claim 1, wherein the homogeneously branched ethylene interpolpolymer is blended with another synthetic or natural polymer.

10 14. The method of Claim 13, wherein the synthetic or natural polymer is an olefin polymer.

15 15. The method of Claim 13, wherein the synthetic or natural polymer is a crystalline polyethylene having a density at 23°C greater than or equal to 20 weight percent as determined using differential scanning calorimetry.

20 16. The method of Claim 15, wherein the crystalline polyethylene has a density at 23°C greater than or equal to 50 weight percent as determined using differential scanning calorimetry.

25 17. The method of Claim 13, wherein the synthetic or natural polymer is a polypropylene.

18. The method of Claim 18, wherein the polypropylene is an isotactic polypropylene polymer.

30 19. The method of Claim 1, wherein the homogeneously branched ethylene interpolpolymer comprises ethylene interpolpolymerized with at least α -olefin.

20. The method of Claim 19, wherein the α -olefin is a C_3 - C_{20} α -olefin.

21. The method of Claim 1, wherein the homogeneously
5 branched ethylene interpolymer comprises ethylene
interpolymerized with propylene.

22. The method of Claim 1 wherein the ethylene
interpolymer comprises ethylene interpolymerized with a
10 styrenic compound.

23. The method of Claim 22, wherein the styrenic
compound is styrene and the interpolymer is an ethylene-
styrene interpolymer.

15

24. The method of Claims 23, wherein the ethylene-
styrene interpolymer comprises from 0.5 to 65 mole percent
styrene, as determined using proton nuclear magnetic
resonance analysis wherein

20

(a) sample preparation is in 1, 1, 2, 2-
tetrachloroethane- d_2 (TCE- d_2) and

(b) spectra are accumulated on a Varian VXR 300 unit
with the sample probe at 80°C and referenced to the residual
protons of TCE- d_2 at 5.99 ppm.

25

25. The shaped interpolymer of Claim 1 or 3 in the form
of film.

26. The shaped interpolymer of Claim 1 or 3 in the form
30 of fiber.

27. The shaped interpolymer of Claim 1 or 3 in the form of a molding.

28. The shaped interpolymer of Claim 1 or 3 in the form
5 of a thermoform.

29. The shaped interpolymer of Claim 1 or 3 in the form of a woven or nonwoven fabric.

10 30. A personal hygiene item comprising the shaped interpolymer of Claim 1 or 3.

31. The item of Claim 30 wherein the item is a disposable diaper.

15

32. The item of Claim 31 wherein the diaper comprises a backsheet or a topsheet comprised of the shaped interpolymer.

33. An infection control item comprising the shaped
20 interpolymer of Claim 1 or 3.

FIG. 1

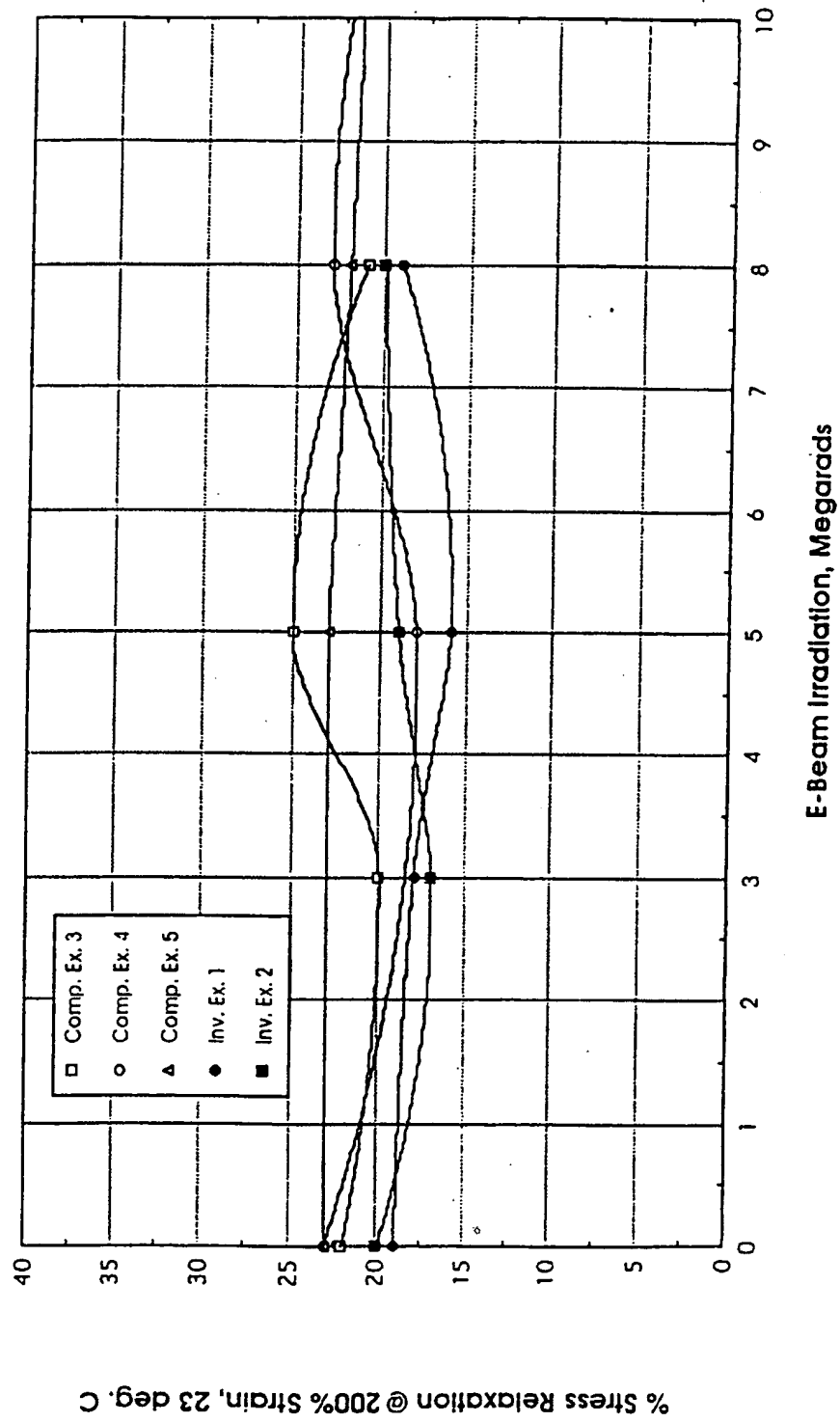


FIG. 2

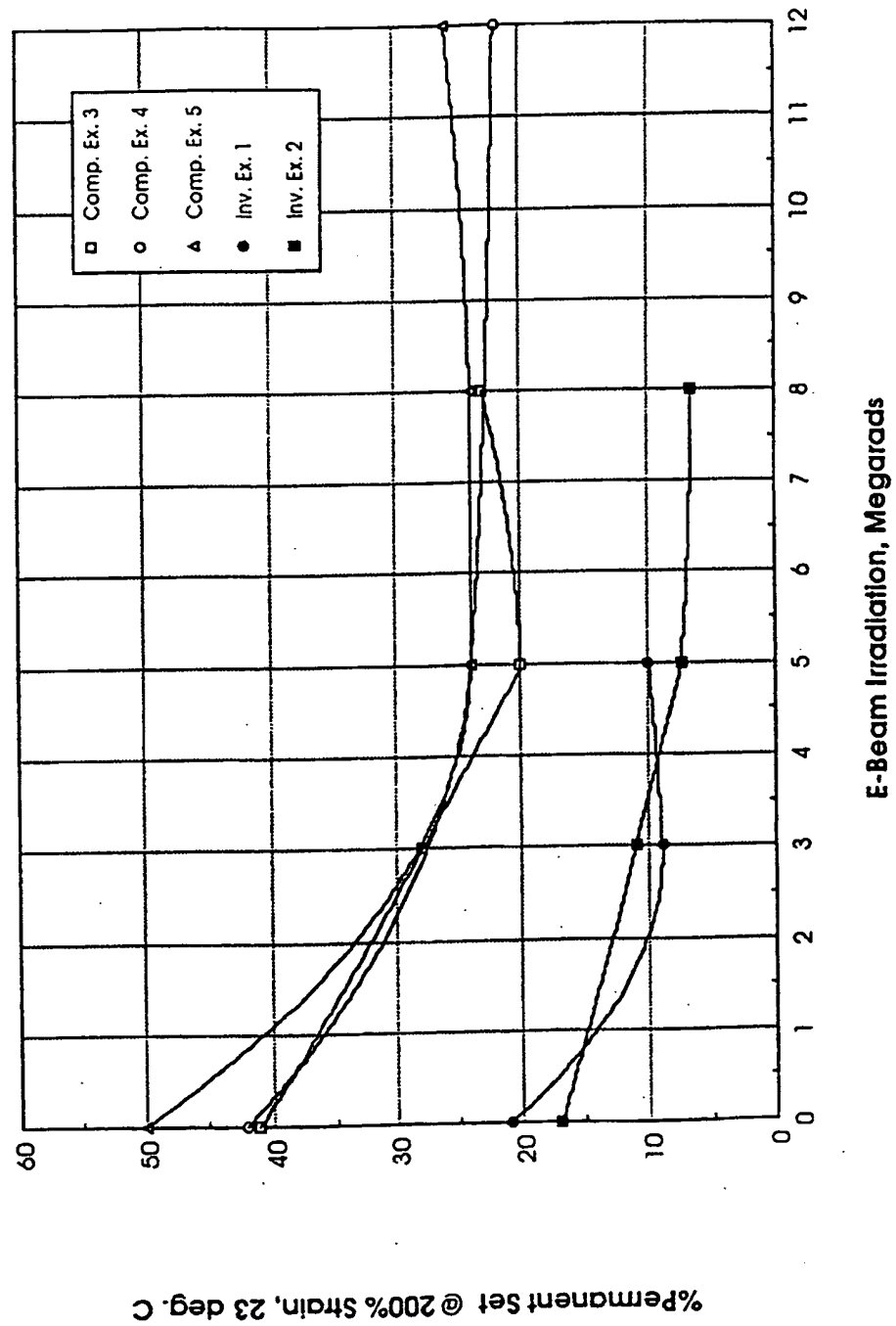
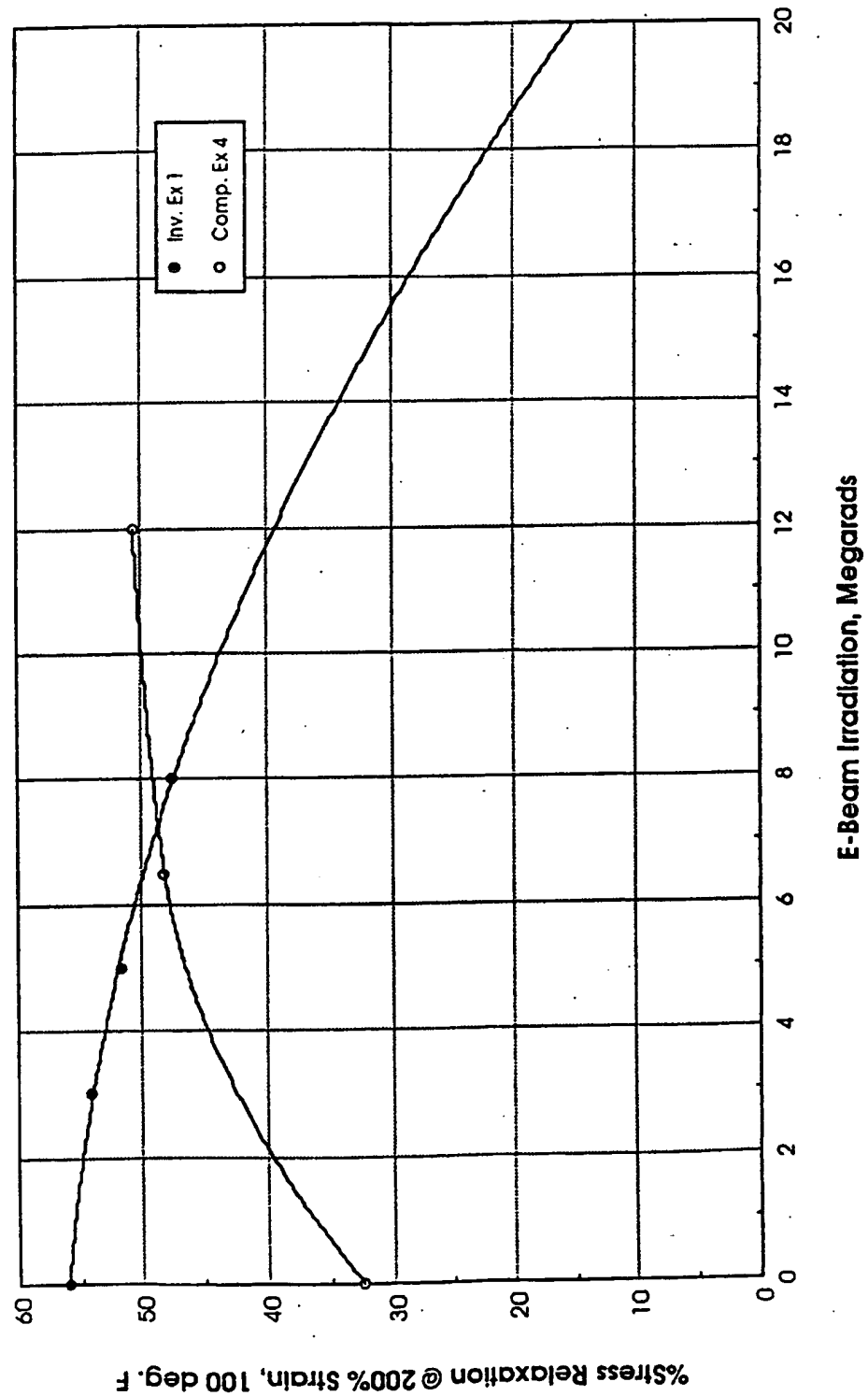


FIG. 3



INTERNATIONAL SEARCH REPORT

International Application No.

PCT/US 99/11745

A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C09J151/06 C08L23/16 C08K5/16

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C09J C08L B32B C09D C08F C08K C08J B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
P, X	WO 98 26001 A (DUPONT DOW ELASTOMERS L L C) 18 June 1998 (1998-06-18) * page 4, line 9-14 ; page 13, line 22 ; page 4, line 35 - page 5, line 13 ; page 7, line 11-35 ; abstract ; page 11, line 20-30 * page 10, line 5-9	1-3,5,6, 8-30
X	WO 97 12919 A (DOW CHEMICAL CO ; FARAH HANI (US); LAUGHNER MICHAEL (US); HOFMEISTE) 10 April 1997 (1997-04-10) * page 27, line 1-23 ; page 46, line 5-12 ; page 81, line 4-5 ; claims 7-9 ; page 49, line 26 - page 50, line 20 * page 9, line 1-12	1,6,8, 11, 13-23, 25-29
	-/-	

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
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- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

11 August 1999

Date of mailing of the international search report

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INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 99/11745

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT		
Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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Y	<p>WO 94 25648 A (DOW CHEMICAL CO) 10 November 1994 (1994-11-10)</p> <p>* page 4, line 6 - page 6, line 25 ; page 8, line 1-10 *</p> <p>page 11, line 9-18</p>	1-33
Y	<p>WO 98 20065 A (DSM NV ; GIJSMAN PIETER (NL)) 14 May 1998 (1998-05-14)</p> <p>* page 6, line 31-36 *</p> <p>abstract; claims 1-16</p>	1-33
A	<p>EP 0 346 862 A (PHILLIPS PETROLEUM CO) 20 December 1989 (1989-12-20)</p> <p>* page 3, line 17-18 ; page 3, line 40-49* page 4, line 24-36</p>	1-30

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Information on patent family members

International Application No

PCT/US 99/11745

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